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पुस्तकालय

विषय संख्या 505
पुस्तक संख्या L84E
V.21
आगत पंजिका संख्या 1

पुस्तक पर किसी प्रकार का निशान लगाना वर्जित है। कृपया १५ दिन से अधिक समय तक पुस्तक अपने पास न रखें।

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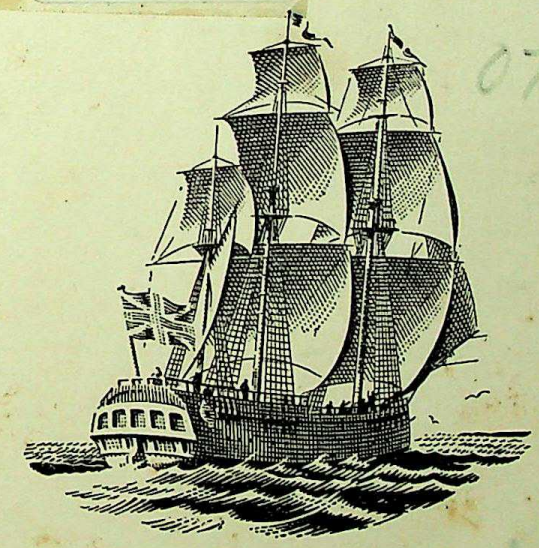

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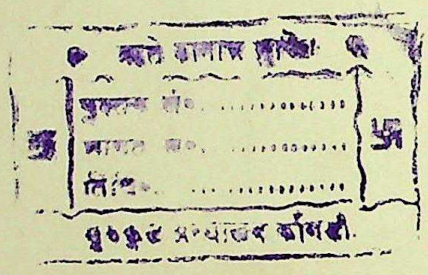

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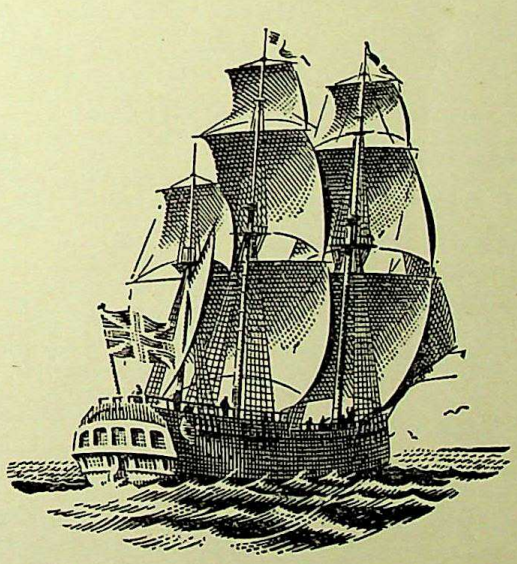

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The jubilee of the nuclear atom

In May 1911, Professor Ernest Rutherford, as he was then, published in the *Philosophical Magazine*—at that time the agency through which many great discoveries were first made known—a paper entitled 'The Scattering of α and β Particles by Matter and the Structure of the Atom'. This paper was the announcement of the nuclear structure of the atom, a seed from which has grown so significant a part of modern physics, including much that can be truthfully called world-shaking. The appearance of the paper, however, caused so little stir that *Nature* did not refer to it; it can be said that in the world of physics there was no recognition that it had any interest except for the comparatively few who were curious about the scattering of alpha and beta particles—a very specialized subject.

Rutherford's paper was, in fact, the culmination of a series of experiments on the small changes of direction experienced by the alpha particles of a fine beam during passage through a thin sheet of matter. Geiger, working under Rutherford's inspiration, had examined the scattering of alpha rays by very thin metal foils and had found that, while most of the particles passed straight through, a small proportion was deflected through angles of up to ten degrees or so. This could be explained in a general way by the laws of probability, but Geiger and Marsden in 1909 found that a very few of the particles incident on the metal foils came right back, emerging on the side by which they had entered the foil.

To most men of science this might not seem to be anything but a chance anomaly, of no special significance. To Rutherford, however, it was an event of amazement. As he said many years later, 'That was a most surprising result, considering the minuteness of atoms and the great swiftness and energy of the alpha-particles which were fired at them. It is just as surprising as if a gunner fired a shell at a single sheet of paper and for some reason or other the projectile bounded back again'. To explain it, he put forward the theory that practically the whole mass of the atom was concentrated within a sphere having a radius about a ten-thousandth of that of the atom itself, as usually estimated, and strongly charged. In the paper of 1911, the charge of the central particle, later called the nucleus, could be positive or negative, the electricity of opposite sign being distributed through the volume ordinarily thought of as the atom. A positive particle could be equally well deflected by a nucleus of either sign. In the case of a negative nucleus it would pass round it and back, as a comet travels round the Sun, and in the case of a positive nucleus it would be repelled before reaching it. A little later, Rutherford decided that the charge must be positive.

Round about 1911, speculations concerning the structure of the atom were not, perhaps, taken as matters about which any degree of certainty could exist, but rather in the spirit in which speculations concerning the habitability of other worlds are taken today. Even Rutherford himself did not, it would appear, fully appreciate the significance of what he had done, and it may be that it was with this in mind that he wrote to Geiger in 1932, 'They were happy days in Manchester, and we wrought better than we knew'.

The birth of the nuclear atom has been recently celebrated at its birthplace, Manchester, in a Rutherford Jubilee International Conference, a pleasant feature of which was the presence of a large number of famous physicists from abroad, including Niels Bohr. Rutherford's laboratory was always a centre of attraction for foreign workers concerned with research into the mechanism of the atom. As early as 1905, for instance, Otto Hahn went to Montreal to work under Rutherford at McGill University. In the spring of 1912, Bohr came to his laboratory at Manchester. It was Bohr who first showed the wider and more precise possibilities of the nuclear model. Rutherford, as has been said, was concerned only with the explanation of the scattering of swift particles by matter: not only were the alpha and beta particles specifically named in the title of his epoch-making paper, but in his great book *Radioactive Substances and their Radiations*, published in 1913 with a preface dated October 1912, there are only two references to the nuclear atom, both concerned with this scattering. There is no suggestion that optical or chemical properties are capable of, or need, explanation by an atomic model.

By applying Planck's quantum theory to Rutherford's general scheme, Bohr showed how the spectra of simple atoms could be quantitatively expressed, the exact value of Rydberg's constant, which governs the frequency of the lines of all the series of the hydrogen spectrum, being deduced theoretically. The immense significance of Planck's theory itself was not recognized at the time: it was not until 1918 that Planck was awarded the Nobel prize for work done at the very

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beginning of the century, and not until 1926 was he elected a Foreign Member of the Royal Society, in the same year as Bohr. Bohr's work did much to make the fundamental significance of Planck's innovation realized. Bohr's first paper was published, in July 1913, in English in the *Philosophical Magazine*, like Rutherford's paper, and was communicated by Rutherford. A second paper of prime importance, in which the electronic structure of the atom was further developed, appeared two months later. It was Bohr whose work, eyed somewhat sceptically at first in many quarters, demonstrated that the nuclear atom had opened a new world of theoretical physics. He not only outlined an electronic structure of the atom and showed how the fundamental facts of line spectra could be explained by it, but directed attention to the application of the electronic model to chemistry. Just as the foundation of the Royal Society has been considered as taking place both in 1660 and 1662—the two hundred and fiftieth anniversary was celebrated in 1912 and the three hundredth in 1960—so the year 1913 may be coupled with 1911 as the date of origin of the nuclear atom.

Recognition of the importance and originality of Bohr's work does not detract from the merit of Rutherford's monumental discovery. Geiger has recorded how, at the time of the discovery, Rutherford 'came into my room and told me that he now knew what the atom looked like', and it was this faculty of seeing what things looked like, this power of graphic simplification, that was characteristic of Rutherford's genius. He saw and he knew.

After Bohr's fundamental advance, the theory of the nuclear atom developed rapidly. Kossel's work during the years of the First World War, showing how both the fundamental features of the X-ray spectra and of chemical valency could be expressed in terms of the electronic structure, was of great value. The appearance in 1919 of Sommerfeld's *Atombau und Spektrallinien*, a work which went through many editions in German and was widely read in English translation, may be considered to indicate that the nuclear atom, born in 1911, had by then outgrown its childhood.

The period before the First World War in which the theory of the nuclear atom was born was, of course, another age, contrasting strangely with the present epoch. The university laboratory, in particular, was a very different place: research was not, for the younger workers, a profession with innumerable openings, and the professor had much undisturbed leisure in which to think. Rutherford in Manchester had little to distract him from his passionate interest in problems of physics, in particular from his enthralled curiosity in all that concerned radioactivity and the atomic complexities connected with it. His academic duties were conscientiously accomplished, but took up only a small fraction of his time. He could keep a particular problem constantly before him. The conception of the nuclear atom was simple, but such fundamental simplifications occur only to minds that have been long and uninterruptedly bent to a complex and apparently intractable subject. The laboratory was his paradise: he said to Harold Robinson, 'Robinson, you know, I am sorry for the poor fellows that haven't got labs to work in.'

Bohr has recorded that Rutherford, speaking with pleasure of his appointment as Cavendish Professor of Physics at Cambridge, expressed fears that the many duties connected with the post would not leave him with the opportunities for research that he had enjoyed at Manchester. It was inevitable that his direct concern with experiment should not be so strong as it was before the war, but under him at Cambridge the progress in research connected with the nuclear atom was notable for great discoveries, the sequel to his long cogitations. In his Bakerian Lecture in 1920, he had foretold the existence of heavy hydrogen and of the neutron. At Cambridge the transmutation of the atom was achieved. To the end, Rutherford retained his extraordinary power of inspiring those who worked with him, in spite of many distractions. But distractions there were.

Today, after another war, things have moved much further from the old traditions that prevailed fifty years ago. The heads of some great university laboratories have little time for direct contact with their research men. They are expected to attend conferences, often lengthy, at home and abroad; to sit on committees concerned with questions often not entirely scientific; to answer a diversity of questions from administrative heads; to concern themselves with building and other projects; and so on. If they take this course they will have, as time goes on, less and less of the undisturbed leisure that Rutherford enjoyed in the great days of the Manchester school of physics from which the nuclear atom and other great, if lesser, discoveries came forth. The new system is, no doubt, evidence of progress and is held by some to be a sign of efficient organization. There might, however, be advantages if it were possible for some of our present-day Rutherfords to spend a substantial part of their time in thinking about fundamental questions.

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The history and relationships of the world's cottons

SIR JOSEPH HUTCHINSON

The great economic importance of cotton needs no emphasis. Annual production of raw cotton is about seven million tons, and it is produced in North and South America, Africa, Asia, and, to a limited extent, Europe. Despite the very wide geographic range of the crop, genetic studies indicate that only four species have contributed to the great diversity of modern cultivated cottons. This article includes a discussion, based on genetic and archaeological evidence, of various theories of how cotton became established as a crop plant.

Students of the cotton plant enjoy the enormous advantage of a comprehensive work of reference: in 1907 Sir George Watt [27] set out all that was then known of the botany of the plant and much of its agricultural history. Modern work on the crop began with S. C. Harland's analysis [11] of the nature of the species distinctions among the cultivated cottons, and led to studies of their origin and evolutionary history [15, 19]. Harland's work was, in addition, a major contribution to genetics.

Harland's study of the genetic nature of the species distinction led to the acceptance of four, and only four, species to embrace the whole vast diversity of the cultivated cottons. Between these four the primary distinction is in chromosome number. Two species, *Gossypium herbaceum* and *G. arboreum*, are diploid, with $n=13$ chromosomes; they are confined to tropical and sub-tropical regions of the Old World. Two species, *G. hirsutum* and *G. barbadense*, are tetraploid, with $n=26$ chromosomes, and it has been shown that, of this complement, half is homologous with the complement of the Old World diploids, and half with the complement of species of the genus growing wild in the New World. The tetraploid species were confined to the New World until they were spread, along with other New World crop plants, over the Old World in recent times.

These four species all bear lint hairs on the seed. The wild species of the genus also bear hairs on the seed, but these are short and scanty, and of a type that could not be spun. The term 'cotton' is properly restricted to the lint-bearing species.

The commercial cottons of the present day are almost all short-term annual plants. They belong to a genus of perennial shrubs, and the primitive forms of the cultivated species are perennial shrubs; the annual forms have been bred and brought into use only in historic times. For a study of the development of the cottons, therefore, the primitive perennials, both wild and cultivated, are the natural starting-point. Primitive perennial types of the cultivated cottons are not difficult to find. The modern textile industry, consisting of great mills dependent for supplies on the cultivation of fairly uniform types over large areas, is of very recent origin, and in all the older cotton-growing countries the remnants of the old textile crafts are still to be found. The craftsman—spinning, weaving, and dyeing the cloth in traditional ways and on a small scale—draws his raw material from small plots, or occasional plants grown in gardens or house compounds. It was from these relics of village crafts that the primitive forms of the world's cottons were collected. They are to be found over wide areas in India and south-east Asia, in the Sudan and West Africa, and in the West Indies, Central America, and northern South America. They occur less commonly in the Pacific islands and even in northern Australia.

These cottons readily run wild and establish

SIR JOSEPH HUTCHINSON
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Was born at Burton Latimer in Northamptonshire in 1902 and was educated at Ackworth and Bootham Schools and St John's College, Cambridge. He worked first at the Cotton Research Station, Trinidad, and then at Indore in India. He returned to the Cotton Research Station, Trinidad, and travelled extensively in the West Indian region. In 1947, he went to Africa and pursued his studies of the cotton crop in the Sudan. In 1949 he became Director of the new Cotton Research Station at Namulonge in Uganda, and in 1957 he was appointed Drapers' Professor of Agriculture in the University of Cambridge. In addition to numerous research papers, he and his colleagues published 'The Evolution of *Gossypium*' in 1944, and he has recently supplemented this with 'The Application of Genetics to Cotton Improvement'.

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themselves in waste places and hedgerows, and on the margins of cultivated land. Watt and other early writers regarded many of them as truly wild, and ancestral to the cultivated cottons. In collecting, it became apparent that there was no line of demarcation between the wild plants of waste places; the occasional plants of house-compounds and gardens that grew from carelessly scattered seeds but which were regularly harvested; and the true crop plants of neighbouring fields. It then became important to decide whether the wild plant was the original from which the cultivated form was developed, or whether it arose as an escape into natural or semi-natural vegetation from cultivated lands.

As our knowledge increased, so the number of forms that could be regarded as independent of human influence diminished. A. Chevalier [4] stated that, of the cottons now to be found in West Africa, none occurs in undisturbed vegetation, and all show signs of having reached their present habitat as cultivated plants. In the Sudan and Ethiopia [21], it is also clear that the cottons exist in association with man, and not wild in natural vegetation. Moreover, in no locality in India are the uncultivated forms of cotton to be found growing in anything like natural vegetation. They have, in fact, all the characteristics to be expected of the abandoned relics of the crops and crafts of an earlier culture.

The situation is similar in the New World. S. G. Stephens [17], collecting cottons in Central America, found that it was useless to look for material anywhere but in villages and on village lands. In the West Indian islands also, the cottons are to be found growing as commensal plants, preserved and harvested for domestic use even if not deliberately sown. There are a few reports of New World cottons now growing wild in natural vegetation. J. B. Hutchinson [13] has given an account of the '*algodón brujo*' of the Salinas de Cabo Rojo in Puerto Rico, and O. F. Cook and J. W. Hubbard [5] recorded the existence of the plant they described as *G. morrilli* on coastal dunes beyond the limits of agriculture in north-western Mexico. The success of primitive types of cotton in establishing themselves on waste land is such that it would be expected that they would sometimes spread into undisturbed natural vegetation, and Stephens [17] has given an account of the transition that is to be observed in Yucatan from domestic cottons to the ecotype that is established on the coastal sand dunes of the Progreso area; there, considerable specialization to the exposed maritime situation has taken place.

In South America also, accounts by E. K. Balls [19] indicate that the cottons found growing outside the present limits of cultivation are escapes and not truly wild plants. Even in the Galapagos Islands, the evidence from both herbarium material and from accessions established in the living collection is that there is a continuous range of form, from those closely similar to cultivated types in western South America to types evidently closely adapted to survival in the wild.

The evidence for the view that types of cotton now found wild are escapes from cultivation is so widespread, and the number of forms for which an origin by escape from cultivation cannot be demonstrated is so small, that in 1947 J. B. Hutchinson, R. A. Silow, and S. G. Stephens [15] set out to account for the origin and development of the cottons on the assumption that the whole process had gone on under domestication. This extreme view is no longer tenable [18], but truly wild cottons are so rare that the differentiation of the modern cultivated cottons is best considered in terms of the cottons that are now, or recently have been, cultivated. Consideration of these early cultivated cottons leads directly to the study of the origin and spread of the annual cottons. In fact, the success of the cottons as crop plants has depended very largely on the comparative ease with which annual types have been selected from the original perennial shrubby forms. The branching pattern of the cotton plant, with vegetative branches arising from the lower part of the stem and fruiting branches from the upper, lends itself to the development of annual forms. By selection of plants giving rise to fruiting branches low on the main stem, the vegetative period of the plant can be reduced, and flowering brought forward far enough to allow a crop to be matured on a spring-sown plant before the onset of cold autumn weather.

COTTONS OF AFRICA AND ASIA

Perennial cottons are confined to areas where there is no winter frost, since all cottons are frost-susceptible. Though they can survive a long dry season, the development of extensive commercial crops in areas of seasonal drought has been much more successful with annual cottons. Consequently the development of annual types has made possible vast extensions of the area in which the plant can grow, and the sub-divisions that have

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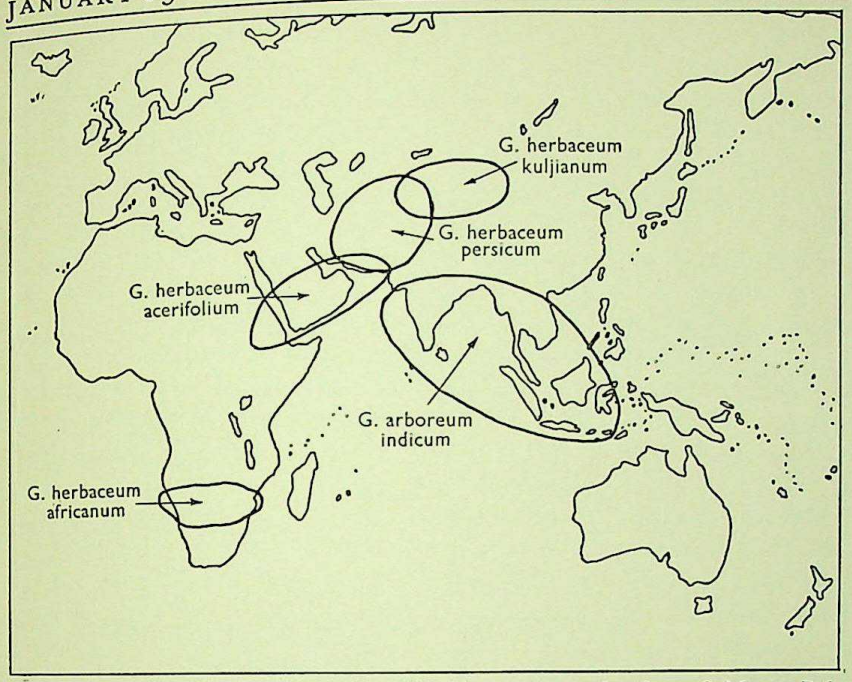

FIGURE 1 – *Distribution of the Old World cottons at the time of Marco Polo (thirteenth century).*

arisen in each of the cultivated species are best described as geographical races. In the cottons of the Old World, annual forms were first developed in *G. herbaceum* (figure 1). In this species the most primitive, and only truly wild, form is race *africanum*; this is a perennial shrub found in the southern African bushveld across a belt of country from Mozambique to Angola and south-west Africa [18]. The most primitive cultivated forms are those found as occasional perennial plants in fields and gardens in Ethiopia [21], in southern Arabia, and on the coasts of southern Baluchistan; these are included in the race *acerifolium*. Inland in Iran, a characteristic group of annual forms has arisen, named race *persicum*. Further north, where the summers are shorter and the winters colder, the species has given rise, by progressive adaptation to shorter seasons, to the *kuljianum* cottons of Central Asia that will mature a small crop in three months from sowing. The annual *persicum* cottons from Iran spread into western India, and provided the first annual cottons in Indian agriculture. The introduction can be dated, since it has been shown [19] that when Dr Hove collected in Gujerat in 1787, the annual form was new to the country, where it provided an alternative to the perennial *G. arboreum* known as Rozi.

The history and relationships of the Indian species, *G. arboreum*, were somewhat more difficult to unravel, since the greater part of the development of the species took place under the influence of an important and expanding textile industry in a country where communications were comparatively good and exchange of seed was easy and frequent. Thus, on the one hand, the geographical races are not so well separated or so distinct as in *G. herbaceum*, and on the other, there has been a much greater development of agricultural forms adapted to particular regions and specific needs.

Silow [23] showed that race *indicum* of peninsular India was more closely related to the cottons of the species *G. herbaceum* than are other races of *arboreum*. Race *indicum* includes both perennial and annual forms; there is good evidence that the perennial forms are primitive. The Reverend E. Terry, who accompanied Sir Thomas Roe on his mission to India at the beginning of the seventeenth century, reported that 'those shrubs bear that Wool three or four years ere they supplant them' [27].

The cottons of northern India and Pakistan are distinct from those of peninsular India. Perennial forms are still to be found occasionally in remote places in Rajputana, and in those parts of the Ganges valley where cotton is not grown commercially but is appreciated as a useful plant for household purposes. This northern form spread widely both eastward and westward. Perennial forms in Assam and Burma are derived from it, and westward it has provided the material out of which the common Old World perennial cotton of Africa has developed. The African form, now known as race *soudanense*, was probably the cotton grown by the people of Meroë, an ancient Nubian kingdom, who were the first in Africa to spin and weave cotton [16].

The selection of annual types in the *arboreum* cottons was no more difficult than it had been in the *herbaceums*, and with the expanding trade of the nineteenth century annual forms of *arboreum* were developed; these not only supplanted their perennial ancestors, but spread into areas where cotton growing had formerly been unimportant. The new overseas trade in cotton and cotton goods brought with it changes in demand also. Manchester had supplanted the Indian village

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artisan as the supplier of the great bulk of cotton cloth. A large export trade in raw cotton grew up, but it was to countries further east rather than to Manchester, and the export demand was for quantity rather than quality. It happened that the annual types selected in the Ganges valley from the northern *arboreums* possessed a higher 'ginning out-turn' (proportion of lint to seed) and a lower quality than those selected from the *indicums* of peninsular India. These northern annuals were sufficiently distinctive to be regarded as a geographical race (*bengalense*), and they spread south into the peninsula at the expense of the *indicums*. Thus there arose a problem that was to beset the Indian cotton industry for many years. Quantity drove out quality, and by misclassification, adulteration, and other malpractices, the reputation of quality-producing areas was ruined, and coarse high-ginning types were spread. Only with the establishment of the Indian Central Cotton Committee in 1921 and the control of cotton movements under the Cotton Transport Act was the situation brought under control and the better-quality cotton protected.

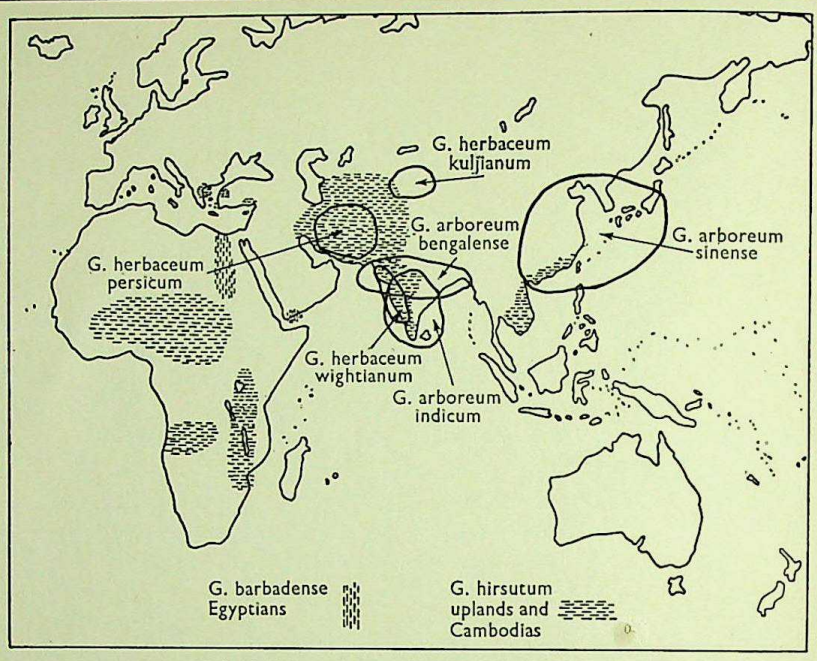

FIGURE 2 – *Distribution of annual cottons in the Old World in* 1960.

The development of annual *arboreum* cottons was not confined to India. They were selected independently in Burma, though they do not appear to have become so widespread there as in India. In south-east Asia, colonists from peninsular India brought perennial cottons with them, probably of the *indicum* type, and in Indonesia some annual types have arisen. But the great development of annual cottons in the Far East was in China (figure 2). According to Watt [27], cotton was grown in Chinese gardens as an ornamental shrub towards the end of the seventh century; cultivation as a field crop began in the eleventh century. The Chinese must have been among the first to develop annual forms of *arboreum*, since most of their crop is grown in areas where winter cold would preclude the growing of perennials on a field scale. The Chinese *arboreums* of the present day—race *sinense*—are the earliest-fruiting forms in the species, and have developed a distinctive character that marks them off from the two main Indian types, *bengalense* and *indicum*.

COTTONS OF THE NEW WORLD

Cotton was grown and used extensively in South and Central America when the New World was first discovered (figure 3). It is impossible now to assess adequately the relative importance of the southern and the central regions. From the coastal strip in Peru, a great wealth of cotton fabrics of outstanding design and technical competence has been recovered, preserved by the dry soil of the desert graveyards. From the moister climates of Central America, nothing comparable has survived, though it is known from the historical records that cotton provided clothing for the people. As in the Old World, so in the New; the ancient cottons were perennial shrubs. Perennials of the South American *G. barbadense* spread throughout the continent as far south as northern Argentina, but apart from a Brazilian form with large leaves, large flowers, and large bolls and seeds fused into a kidney-shaped mass, there are no well-defined geographical races among the perennials of *G. barbadense*.

Differentiation among the perennials of *G. hirsutum* is much more pronounced. Two important perennial races were early recognized. One of these, race *marie-galante*, includes the largest cottons of all. On the coasts of the Spanish Main, from Panama to Trinidad, and thence north to the Greater Antilles and south to northern Brazil, very large shrubs or small trees up to 20 ft in height are to be found in house-yards and in

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small cultivations, and sometimes wild among dry coastal vegetation. These were the basis of the early cotton cultivations of the West Indies, and the imprint of human selective forces is still to be detected in their modern descendants [26]. Outside the Americas, race *marie-galante* is known only from Ghana [16], where it was introduced from the West Indies by the Basle missionaries.

The second important perennial race of *G. hirsutum* is race *punctatum*, a much smaller, bushy cotton, to be found round the coasts of the Gulf of Mexico from Yucatan to Florida and the Bahamas. It has been recorded infrequently from Cuba, Haiti, and Puerto Rico, but not from Jamaica or the Lesser Antilles. Race *punctatum* was introduced into West Africa about the end of the seventeenth century. There it was very successful, and spread across the continent south of the Sahara, completely replacing the Old World cottons as a raw material for the local spinning and weaving industry. It was also introduced into the island of Bourbon (Réunion) by the French, and taken thence to the Malabar coast of India by the East India Company. In the 1930s it was still to be found mixed with a perennial form of *G. arboreum* race *indicum* in the Nadam crop of Madras. British settlers took it to northern Australia; their settlement was a failure, but the cotton survived, and was collected about 1930 from the neighbourhood of Port Essington.

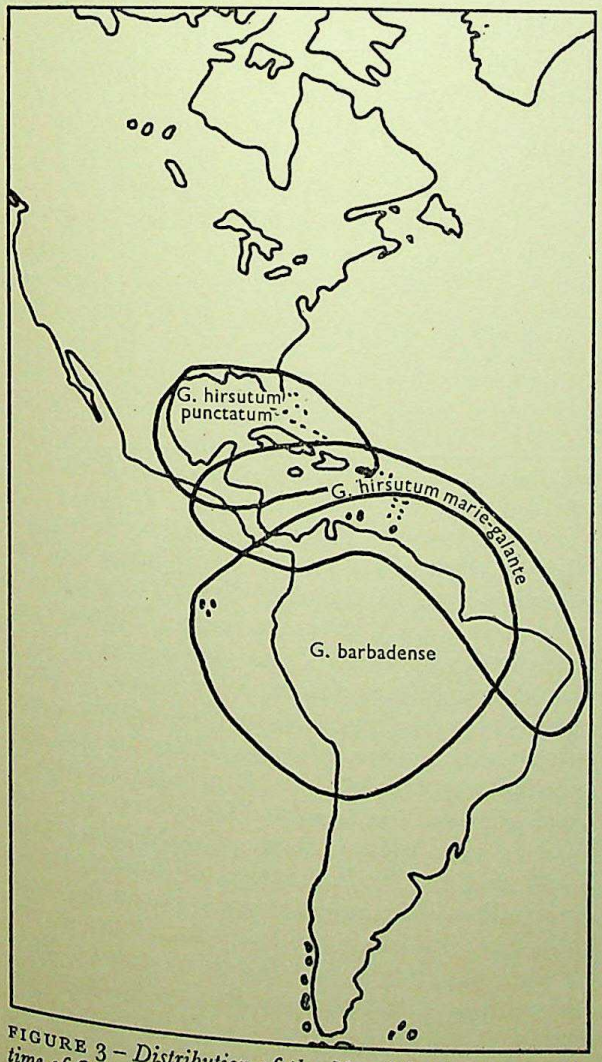

FIGURE 3 – *Distribution of the New World cottons at the time of Columbus.*

Between 1945 and 1948 three expeditions collected extensively in the centre of origin of *G. hirsutum* in Central America. Apart from a very few, apparently introduced, forms of *G. barbadense*, all the cottons collected were forms of *G. hirsutum*. This material revealed a geographical differentiation formerly unsuspected. Northern Guatemala and Yucatan were occupied by typical race *punctatum*, with the specialized form collected by Stephens on the coastal sand dunes near Progreso (see above)—race *yucatanense*—representing the escape of an exceptionally specialized form into natural vegetation. The second well-known race, race *marie-galante*, was represented in collections from Salvador and southern Guatemala. These plants, though evidently belonging to *marie-galante*, were smaller bushes than the large forms of the Spanish Main and the Lesser Antilles. They still possessed the characteristic dominance of the main stem that makes the development of tree forms possible, and distinguishes *marie-galante* from the bushy, much-branched *punctatum*.

In central Mexico in Oaxaca, Puebla, and Morelos the perennial cottons were bushy forms with very broad and intensely hairy leaves. These were named race *morrilli*. In the state of Guerrero it appears that a race—*palmeri*—of pyramidal shrubs with deeply dissected leaves and strong anthocyanin pigmentation is almost universal. Last among the perennials is a strong-growing, lax shrub found in a rather small area round the coasts of the Gulf of Tehuantepec.

One other race, *latifolium*, was found in the State of Chiapas in Mexico and in neighbouring regions in Guatemala. This is an annual cotton. It is not an early-fruiting, specialized agricultural type, and it may persist for more than one year. But, as distinct from the perennial races, it gives its major crop in the first year. There are two main forms: a small-fruited form that occurs throughout the area, and a large-fruited, large-leaved form found in the vicinity of the town of

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Acala in the State of Chiapas in Mexico. This race appears to be the foundation stock of all the annual *G. hirsutum* cottons. In Central America it has spread into the territories of races *morrilli* and *palmeri*. There, the advantage of annual cottons for commercial cultivation has led to the spread of the neighbouring annual form, and natural hybridization with the local perennial has led to the emergence of an annual with some of the characters of its perennial predecessor.

The spread beyond Central America has been enormous (figure 4). First came the colonization of the Cotton Belt of the United States of America. The source of the first introductions is not known, but there are numerous records of subsequent introductions from Mexico, at intervals down to the time of the discovery of the big-bolled types from Acala which have given the modern very high-yielding commercial types of that name now grown in Arizona and California.

The annual *hirsutum* cottons of the United States became known as 'Uplands' (to distinguish them from the 'Sea Island' cottons of the coastal islands, which belong to *G. barbadense*), and their development coincided with the development of the cotton industry in Lancashire. As an index of the rate of growth of the crop, in 1784 Customs officers in Liverpool seized eight bags of cotton on the ground that so much could not have been produced in America. In 1912–13 Great Britain consumed over three and a quarter million bales of American cotton.

The natural desire of other countries to share in the wealth of this export trade, together with such special stimuli as the cotton famine in Lancashire caused by the American Civil War, led to the introduction and trial of Upland cotton in most tropical and sub-tropical countries of the world. The modern Russian crop in Central Asia is composed of Upland types. Apart from the Nile Valley, all cottons grown in Africa are now Uplands. In India and Pakistan, Upland cottons were first introduced at the time of the American Civil War. They were a dismal failure, owing to the attacks of the jassid pest. Upland cottons went out of cultivation as a crop, but persisted as occasional plants in crops of Old World cottons. Under the shelter of the Old World types, Upland types were not altogether eliminated, but those with some of the leaf hair that gives resistance to jassid attack were favoured. Fifty years later, it was possible for agricultural botanists to select, from the mixed crops, Upland types that had sufficient resistance to be grown satisfactorily in pure stands. This was the beginning of the successful establishment of Upland cotton as a crop in India. A different source of American-type cottons for use in India was discovered in the second decade of the twentieth century. Madras cotton-spinners drew the attention of Government botanists to a very useful type of cotton they had been able to import from Cambodia. Seed was obtained, and became the foundation stock of what is now known as the Cambodia crop of South India. Cambodia is a *latifolium* type; its outstanding characteristic is the extremely dense coat of long hairs on the leaf, which gives it very high resistance to jassid. It appears to have been derived from direct introductions by the Spaniards from Central America to the Philippines. The Central American cottons are short-day plants, and when grown in the long summer days of

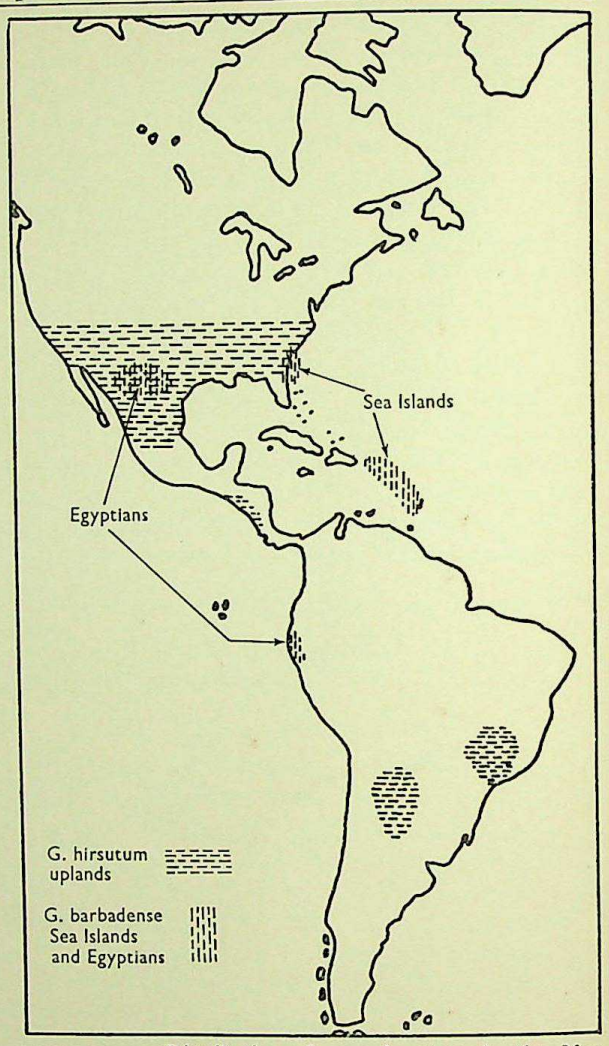

FIGURE 4 – *Distribution of annual cottons in the New World in* 1960.

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northern latitudes they do not fruit until autumn, and consequently they lose their crop when the frost comes. The establishment of the Uplands in the United States depended on the selection of types tolerant of long days and able to mature a crop before the onset of cold weather. Consequently, Upland cottons, wherever they have been taken, are unaffected by day length. The Cambodias, on the other hand, though early fruiting in the short days of Madras, become progressively later-fruiting as they are taken further north, and have in consequence never been successful in competition with Upland types in central or northern India or Pakistan.

The *latifolium* cottons have become the dominant cottons in the world's crop. United States commercial varieties of Upland have formed the basis of the cotton crops of Africa, of most of South America, and increasingly of Central America. Uplands and Cambodias are invading the Chinese crop, and where cotton crops are developed in south-east Asia, they will undoubtedly be based on these types, and on hybrids between them.

The development of annual cottons in *G. barbadense* took place later than in *G. hirsutum*. The *barbadense* cottons of the Sea Islands of South Carolina arose from an introduction from either the Bahamas or Jamaica in 1786. In the Sea Islands only annual cropping was possible, so the type introduced must have been one that was already capable of producing a worth-while crop in the first year. Types are known among the *barbadense* cottons of western South America that fruit early enough, are independent of day length, and are of good enough quality, to have provided a source from which the annual cottons of the Sea Islands might have been derived [14]. The route by which they reached the Carolinas must remain a matter for speculation.

The outstanding character of the Sea Island cottons is their very high quality. Though a wide range of fine cottons is now available in other groups, no other cottons have been bred to the quality of the best of the 'superfine' Sea Islands. Sea Island cotton has always been a small crop. In the first decade of the twentieth century it was introduced into, and became established in, some of the islands of the Lesser Antilles, but the main crop in the United States was virtually extinguished when the boll weevil, spreading across the Cotton Belt from Mexico, reached the eastern seaboard.

EGYPTIAN COTTON

There was a prosperous and growing market for fine cottons throughout the nineteenth and the first half of the twentieth century, and it was largely supplied by another race of annual *barbadense* cottons, the Egyptian cottons. Cotton-growing in Egypt based on *barbadense* cottons was initiated by M. Jumel with perennial types in the years following 1820. The development of the Egyptian crop from this small beginning has been recorded by G. C. Dudgeon [6], and the sources of *barbadense* cottons and the probable route by which they reached Egypt are fairly well understood [16]. Jumel's perennial probably reached the Nile valley from southern Nigeria by way of the trade and slave routes.

In Egypt, cultivation of a perennial crop called for a radical redevelopment of the irrigation system. The old basin system, whereby the land was flooded at high Nile and a crop was grown on the water stored in the alluvial soil, made no provision for watering at low river-levels. Canals were deepened to bring the water to the field margins at low river, and lifts were installed. Cotton proved so successful that it provided both the incentive and the finance for the building of barrages on the river and high-level canals, to provide perennial-flow irrigation. Meanwhile, Jumel's perennial was so successful that other cottons were introduced for trial. None of these were satisfactory except Sea Island types, which were grown on a small scale for some years. Being a form of *barbadense* cotton, Sea Island alone of all the types introduced gave vigorous and fertile hybrids with Jumel's perennial. Out of such hybrids, cotton breeders selected types with the annual habit and some of the quality of Sea Island, together with some of the vigour and cropping characteristics of the old perennial. Thus a new race of annual *barbadense* cottons arose, members of a New World species but bred in, and adapted to, the Nile valley. Furthermore, the replacement of basin irrigation by high-level flow irrigation having been undertaken under the stimulus of the need for water at all seasons for a perennial cotton crop, the perennial plant was replaced by an annual, and the new irrigation system was used to grow two short-term crops each year instead of one perennial.

THE ORIGIN OF CULTIVATED COTTONS

All four species of the cultivated cottons were established crop-plants long before the beginning of historical records. Evidence of their status before the dawn of history is available only from a few specially favourable places. A fortunate

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accident led to the preservation of a small fragment of cotton fabric and a small piece of cotton string in the neck of a silver vessel recovered during excavations at Mohenjo-Daro in West Pakistan. The fragments, which are believed to date from about 3000 B.C., were made from raw material indistinguishable from the product of the indigenous coarse *bengalense* cottons found in the area at the present day. Both fabric and string were well made, and indicated the existence at that time of a well-established, mature textile craft, of which cotton must have been a familiar raw material.

In the New World, the earliest known textiles were discovered first by Dr Junius Bird at a site on the north Peruvian coast known as the Huaca Prieta [2, 3]. The Huaca Prieta is a mound made up of the occupation-refuse of a people who did not use pottery and did not grow maize. They did grow cotton, beans, and some cucurbits. Their cotton was very short-fibred and very coarse, and among present-day cottons is most closely matched by some of the very coarse primitive *barbadense* cottons of the same area. The textile craft of the Huaca Prieta appeared at first sight to be very primitive, but a recent reconstruction of a Huaca Prieta fabric (J. B. Bird, personal communication) has shown an unexpectedly elaborate pattern worked into the material. Evidently, even at that date (*c.* 2400 B.C.), the people of the Huaca Prieta were familiar with cotton, and were far beyond the stage of experimenting with a new raw material.

The use of cotton as a textile goes back far into prehistory in both hemispheres. The archaeological record is inevitably incomplete, since cotton fabrics and cotton plant material have survived only in the driest of the areas in which the crop is cultivated. For further evidence on the origin of the cottons it is necessary to study the botany and cytology of the cultivated species and their wild relatives.

The wild members of the genus *Gossypium* are rare shrubs of the semi-arid and arid regions of the tropics. Watt [27] recorded species from Australia, India, North and Central America, and the Galapagos Islands. Species are now known also from continental South America, Arabia, and Africa; they are listed and described by J. H. Saunders [22]. The wild species do not bear seed hairs that can be spun, and one of the major problems of the origin of the cottons is to account for the origin of lint. The simplest hypothesis is that the four lint-bearing species derived their lint from one common ancestor, and the history of the true cottons will now be reconstructed on this basis.

The chromosome complement in *Gossypium* is basically $n = 13$; all the wild species save one are diploid. The two Old World species of the true cottons are also diploid. The two New World species of cotton and one wild species (*G. tomentosum* in Hawaii) are tetraploid. Of the diploid wild species, the African species of the section Anomala are comparatively closely related to the Old World diploid cottons. There are two species, *G. anomalum* and *G. triphyllum*, and both have been crossed with the two Old World cottons. The hybrids are moderately fertile, and gene transfer and genetic analysis have been carried out successfully [23]. D. U. Gerstel [9] has shown that the chromosome complement of *G. herbaceum* is very closely homologous with that of *G. anomalum*. The complement of *G. arboreum* differs from those of *G. herbaceum* and *G. anomalum* by one translocation. It is in *G. herbaceum*, therefore, that one would logically look for the wild prototype of the diploid cottons. The Anomala are distributed in arid areas on the southern borders of the Sahara and in Angola and south-west Africa. The wild form of *G. herbaceum*, race *africanum*, occurs in natural vegetation right across the dry bushveld and semi-arid tracts of southern Africa from Mozambique to Angola. It occupies, in fact, an area adjoining that of the most closely related wild lintless species, but it has no obvious contact with the cultivated races of its own species. It has all the characters that would be expected of the original wild form: the perennial bushy habit and the hard seeds with a marked period of dormancy adapted to survival in the wild, and a coat of rough, wiry lint on the seeds, long enough and copious enough to be spun if nothing better offered, but of the lowest quality in terms of modern textile materials. The only substantial difficulty in regarding it as ancestral to the cultivated cottons is the enormous distance between the natural habitat of *africanum* and the areas occupied by the primitive cultivated form of the species, race *acerifolium*, in Ethiopia and southern Arabia. If *africanum* is ancestral to the cultivated forms, it must have been carried far to the north at a very early date, or its range must formerly have been vastly greater than it is now. Travel up and down the East African coast has gone on from ancient times, and the trade in gold from the area, in what is now Southern Rhodesia, in which *africanum* grows wild, is of unknown

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antiquity. It seems most likely, therefore, that the early domesticated forms of *herbaceum* arose from *africanum* types carried by travellers northwards to Ethiopia or Arabia.

In considering the origin of the *arboreum* cottons, the most important evidence is that on the morphological similarities and genetic relationships between the perennial forms of *arboreum* race *indicum* and the *herbaceums*. Perennial forms of *G. arboreum* race *indicum* and of *G. herbaceum* race *acerifolium* are extremely difficult to distinguish unless good fruiting material is available, whereas all other forms can be readily and confidently distinguished in all stages of growth. Silow [24] has shown that the genotype of race *indicum* is more closely related to that of *G. herbaceum* than is the genotype of any other race of *arboreum*. The perennials of race *indicum* are native to western India, and the evidence suggests that the species *G. arboreum* arose by differentiation from a primitive *G. herbaceum* stock introduced into western India. Thus, as far as the Old World cottons are concerned, the origin of lint may be traced to *G. herbaceum* race *africanum*, or something closely related to it.

The problem of the origin of the New World cottons is more difficult. They are tetraploid in chromosome constitution, and their chromosome complement is made up of one set homologous with the complement of the diploid Old World cottons and one set homologous with the complement of the wild diploid species of the New World. They are thus allopolyploids, and a related allopolyploid was synthesized by J. O. Beasley [1] from a cross between *G. arboreum* and the wild Arizona species *G. thurberi*. Chromosome-pairing in crosses between the new allopolyploid and New World cottons was good; nevertheless, the synthetic allopolyploid was not a true New World cotton. It was only partially fertile, and the hairs on the seed coat could hardly be regarded as true lint. Subsequently Stephens [15] showed that the Peruvian *G. raimondii* was morphologically considerably nearer to the type one would postulate to give the characters of the New World cottons when crossed with one of the Old World cottons. He was able to produce two hybrid plants, one from the cross *G. arboreum* × *G. raimondii*, and one from the cross *G. herbaceum* × *G. raimondii*, and showed that they did in fact resemble New World cottons more closely than the *G. arboreum* × *G. thurberi* cross. Unfortunately, he did not succeed in inducing polyploidy, and, despite many attempts, it has not been possible to repeat the cross.

Though *G. raimondii* appears to be more nearly related to the New World diploid ancestor of the New World cotton than is *G. thurberi*, it possesses a number of characters that can hardly have been present in the true ancestral type, and it cannot be as close to the New World ancestor of the polyploid cottons as *africanum* is to the ancestor of the diploids.

In view of the difficulty of making crosses from which synthetic tetraploids might be produced, interest turned in the direction of genome analyses of hexaploids, made by doubling with colchicine F_1 hybrids between New World cottons and wild New World diploids. Stephens [15] first reported on the cytology of a *G. barbadense* × *G. raimondii* hexaploid, and showed that pairing at meiosis was such as would be expected if the *G. raimondii* set were rather closely homologous with one set of the *G. barbadense* complement. Gerstel [10] conducted a genetic analysis in back-crosses of hexaploids to New World cottons, and showed that segregation in *G. raimondii* crosses gave ratios much nearer to those expected in autopolyploids than did segregation in *G. thurberi* crosses. Such evidence as is available on other wild New World species indicates that they are no more closely related to the allopolyploid New World cottons than is *G. thurberi* [15].

Since the New World cottons originated from a hybrid between two diploids, one of which was closely related to the Old World cottons, it is not necessary to postulate a separate origin for their lint. On the other hand, since the other diploid parent was related to the Peruvian *G. raimondii*, it is necessary to consider how an Old World diploid and a New World diploid could have come together in western South America so that the cross could take place. The first attempt to account for the meeting of the two parents was by Harland [12], who suggested that they came together on a land bridge across the Pacific Ocean. Little evidence has been adduced for the existence of such a land bridge, and it was later suggested [15, 19] that the link was provided by civilized man, migrating eastward from the Old World and taking his Old World cottons with him. Landing on the Peruvian coast, he would find suitable conditions for his simple agriculture in the valleys of the rivers draining the Andes and crossing the coastal desert to the sea. In those valleys, *G. raimondii* grows naturally, and from natural crosses between the introduced Old World cotton and an ancestor of *G. raimondii* arose allopolyploids from which the New World

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cottons sprang. The hypothesis fitted the known facts, and its usefulness as a working tool was demonstrated when Bird [3] used it to help in the search which led to the excavation of the Huaca Prieta and the discovery of the oldest known textiles in the New World.

An examination of seeds, carpels, and lint from the early Huaca Prieta deposits revealed nothing to suggest the presence of an Old World diploid cotton. Moreover, F. Engel [7] has since shown that habitation sites of the Huaca Prieta type of culture occur over a great stretch of the Peruvian coastline, far beyond the area in which *G. raimondii* now exists.

It may well be, of course, that *G. raimondii* was formerly more widely distributed, or that there were other, related species in the Peruvian valleys. The pressure of the population of coastal Peru on the very limited resources of water and of vegetation is such that the natural flora has been virtually eliminated and other wild species besides *G. raimondii* may have existed in former times. Nevertheless, though restriction of distribution and even extinction may have affected the situation greatly, it cannot be claimed that evidence for the origin of the allopolyploid cottons in coastal Peru has increased greatly since the hypothesis was put forward, and alternative theories must now be examined.

G. L. Stebbins [25] has suggested that the diploid Old World parent reached the New World by way of China and Alaska, and the possibility of an Antarctic route has been proposed. Both the northern and the southern routes are advocated on the ground that at one time mesophytic temperate woodland floras existed that were common to the two hemispheres. *Gossypium* is a genus of xerophytic perennial shrubs adapted to the arid tropics. No member of the genus would grow in an ecological situation where temperate woodlands existed, and until the modern development of short-term annual cottons under domestication, no member of the genus would survive in a climate with winter frosts. It is therefore reasonable to conclude that contact between Old World and New World species did not come about by migration round the Pacific, either by a northern or by a southern route.

The remaining alternative to a trans-Pacific link is a link across the Atlantic, and Gerstel [9] has recently suggested that this should be considered. He showed that in *G. herbaceum* the chromosome order does not differ substantially from that in the wild African *G. anomalum*, and he concluded that the *G. herbaceum* order is primitive. *G. arboreum* differs from the primitive order by one translocation, and the corresponding chromosome set in the New World allopolyploids differs from that in *G. herbaceum* by two and from that in *G. arboreum* by three. He therefore argued that *G. arboreum* is not in the main line of development from the primitive chromosome pattern to the modern allopolyploids. Since *G. arboreum* is the cotton of the Indian sub-continent and of the western coasts of the Pacific basin, it seemed to him unlikely that the link between the primitive diploids represented by *G. herbaceum* and the New World cottons of South and Central America was to be sought across south-east Asia.

The difficulty of supposing that the allopolyploids arose following migration of an Old World cotton across the Atlantic lies not only in accounting for the ocean passage, but in understanding how the two parents came in contact after the Old World cottons reached South America. Not only is the closest known New World wild relative confined to the Peruvian coast, but the whole group of New World wild species of *Gossypium* is distributed west of the continental divide. Nevertheless, certain other fragments of evidence seem to point towards an eastern South American origin for the cottons. Engel [8] has reported that the crops of the ancient pre-ceramic cotton-using cultures of the Peruvian coast include groundnuts (*Arachis*), which originated east of the Andes, probably [20] in north-western Argentina, suggesting that the cultivators reached the coast from the mountains, and not from the sea. If they came over the mountains, they presumably brought their cotton with them.

Archaeological evidence from pre-ceramic cultures in Peru is critical. If it could be shown that the people to whom these cultures belonged came there by sea, the recent-origin theory of the development of the New World cottons would be greatly strengthened, and a search of natural vegetation in remote valleys of the Peruvian coast should be undertaken in the hope of discovering a wild diploid species related to *G. raimondii* with characters more nearly those appropriate to the ancestor of the New World cottons. If, on the other hand, it turns out that the earliest Peruvian cotton farmers reached the coast from the mountains, the wild American diploid ancestor of the New World cottons might be sought east of the Andes, and not on the Pacific slopes. One would naturally look in arid areas for such a

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plant, and in the regions in which the other crops of the early cotton users originated.

To accept an eastern South American origin for the New World cottons would involve accepting also an ancient and not a recent origin for the allopolyploids. The cultivation of cotton on the Atlantic coasts of Africa is very recent, and it would be necessary to suppose that the parents of the allopolyploid were both wild and came in contact by natural spread. The present distribution of the wild species of *Gossypium* has been interpreted [15] in terms of the distribution of the major land masses before the continents drifted apart. If the main species distinctions in *Gossypium* were already established at that time, and if a suitable wild diploid existed east of the Andes, a type akin to the South African *G. herbaceum* race *africanum* might have come naturally in contact with its counterpart, and the allopolyploid might have arisen in that way. Then Stebbins's contention would be upheld, that the New World cottons are among the oldest, and not the youngest, of the known allopolyploid species.

The trail of speculation about an ancient Atlantic origin for the New World cottons is as long and as scantily supplied with beacons as that about a recent Pacific origin. Nevertheless, speculation of this kind is worth-while. For example, the original speculation influenced Bird in the choice of the Huaca Prieta as a site for archaeological study. Following Gerstel's suggestion and the archaeological indications reported by Engel, it is possible to set out the conditions governing the possibility of a transatlantic link, and to point to two lines of investigation—archaeological in Peru and botanical east of the Andes—that might make it possible to decide between the two hypotheses. This is a new and unexpected turn in the inquiry into the origin of the cottons. No mention has been made here of the third allopolyploid, *G. tomentosum*, which is a wild fuzzy-seeded—but not linted—species in Hawaii. Ever since the cytological situation in the genus was set out it has seemed that if the origin and relationship of *G. tomentosum* could be elucidated, the problem of the New World cottons would be solved. Now it appears that the origin of *G. tomentosum* is a separate and probably unimportant problem, and the real interest lies in the Peruvian Andes, and in the origin of the earliest cotton farmers of the Peruvian coast.

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The tuatara in its natural habitat

W. H. DAWBIN

The anatomy and embryology of the tuatara, sole living representative of a once widely distributed group of reptiles, have been extensively studied and described. By contrast, little attention has been paid to its habits, partly because of the inaccessibility of the few remaining colonies. This article describes systematic observations of tuataras in their natural surroundings: the observations were made at various seasons of the year, both by day and by night.

The tuatara, *Sphenodon punctatus*—a reptile that may reach 60 cm in length—is confined to a few offshore islands of New Zealand, and has been seen in natural conditions by few people. It has become, nevertheless, one of the best-known reptile species in the world, because of the interest aroused by A. Günther [1] in 1867, when he described material sent to him in London. This showed clearly that, despite the tuatara's lizard-like appearance, it was so different from lizards in skeletal features that it clearly belonged to a separate order; this he called Rhynchocephalia, meaning 'beak-head'. This name refers to the beak-like tip of the upper jaw, separated by a notch from the remainder of the jaw. The main feature that separates this order from lizards is, however, the presence of complete bars of bone bounding all sides of two large apertures or fenestrae in each side of the skull; in lizards, by contrast, the lower fenestra has no border of bone. The ribs have dorsal processes similar to the uncinate (hooked) processes of crocodiles and birds, and the floor of the abdomen contains a bony basketwork, somewhat resembling that of crocodiles but absent from many true lizards. The combination of these and several other skeletal features distinguish the tuatara from all recent reptiles. Furthermore, the tuatara is unique among modern reptiles in that the male has no organ of copulation.

The other three recognized orders of living reptiles include crocodiles and alligators, turtles and tortoises, lizard and snakes. Each of these orders is represented by a range of species, whereas the tuatara is the sole living representative of the Rhynchocephalia, the lone survivor of a once much more widely spread group. Although several species or sub-species have been proposed on the basis of differences of colour and body proportions, there seems to be no real justification for this, as the differences are no greater than have been observed within individual colonies. Fossil remains of other types have been found in various parts of Europe, North America, Africa, and Asia, and many of the skeletons date from the Triassic period of some 200 million years ago. Curiously, there are no fossil 'beak-head' remains known from any country, including New Zealand, throughout the 75 million years of the Tertiary period, so the problems of the route to New Zealand and time of arrival are still subjects for speculation only. However, some of the Triassic beak-heads of the northern hemisphere were remarkably similar to the present-day species, showing how little the tuatara has changed from the earlier forms. The immense period through which the tuatara has persisted without significant skeletal change is one of the most spectacular examples of conservatism in structure known among land vertebrates.

The great interest caused by these findings is indicated by the number of articles that have been written on the tuatara. More than 150 technical, or semi-technical, papers and reviews have been published, of which the most detailed is by O. von Wettstein [2], and very numerous popular accounts have appeared in magazines and newspapers. The anatomy and embryology have been well described from material collected mainly in the nineteenth century, that is, before the justifiably increased protection given to the animal by the New Zealand Government during the present century. All too many of the descriptions of the tuatara's habits, however, have been based on

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brief daytime observations or second-hand reports. Much space would be needed to evaluate and comment critically on all the past work, so that it seems more useful to give here a brief preliminary description of some observations made during recent field-studies than to prepare a further review of the already voluminous literature on the tuatara.

During the 7th Pacific Science Congress, held in New Zealand in 1949, Dr K. P. Schmidt visited Karewa Island [3], which is inhabited by the tuatara, and later he asked me to accompany him on a brief visit to Stephens and Trio Islands in Cook Strait. Stephens Island (figure 1) rises to 1000 ft and is a square mile in area. In good conditions it can be reached by an eight-hour launch journey from the North Island, but it sometimes takes up to three days before it is possible to make a landing. Observations of the tuataras suggested that a long-term field programme might be feasible. In consequence, I have since returned for more than twenty visits of about a week each, through all seasons of the year, accompanied each time by parties of up to fifteen research workers and students. Once ashore, the team has worked throughout the nights, because the nocturnal habits of tuataras are such that daytime observations alone could be just as misleading as night observations alone would be on such phenomena as the flight habits of butterflies.

In order to determine growth-rates in the field, it is necessary to mark animals of varied sizes, and later to compare the increments between the time of marking and recovery. With a known growth-rate, it should be possible to make an estimate of the age of individuals, and then of the age structure of the wild population and of the relative mortality rates at different ages. To this end, more than sixty people have accompanied me on one or more of the twenty study-periods since 1950, and have assisted in capturing the 750 animals that I have measured, weighed, and marked to date. In addition, observations on the life cycle and the variation in seasonal activities were made, and were supplemented during short visits to some of the other islets inhabited by tuataras.

DISTRIBUTION

The total number of islets around the New Zealand coast from which the tuatara has been recorded is twenty. They occur in two separate groups, one between North Cape and East Cape of the North Island, and the other in Cook Strait between the North and South Islands. Nearly all the islets are wholly or partly cliff-bound and subject to rather rapid changes of weather. Sea conditions often make landings hazardous or impossible, and in consequence only a few of the islands have been investigated by zoologists on more than sporadic short visits. It is partly for this reason that it has taken so long to fill in many details of the tuatara's life history.

Collectively, the tuatara islands can be characterized by a number of features. In undisturbed areas, nearly all have a coastal type of low forest or scrub cover, with a large number of plant species that prefer, or at least can tolerate, salt spray. Under the forest cover, or the grassland of recently modified areas, the ground is normally riddled by the burrows of one or more species of petrels, including shearwaters, prions, and diving petrels. On many of the islands there are also colonies of other marine birds, such as penguins, cormorants, gulls, and terns, so that the total concentration of sea birds during their breeding seasons is enormous; in addition, some species remain resident throughout the year. As a result, the soil is intensively manured and subjected to tremendous overturning by bird activity. Soil acidity, phosphate, and nitrate are all exceptionally high, and this has a direct effect on the species-composition of the plants present. The movement of the birds about the forest floor breaks off many young shoots and seedlings, keeping it much more open than in forests unoccupied by such colonies. Possibly as a result of the high manuring, there is an exceptional number of small cryptozoic ground organisms and very high concentrations of ground insects such as wingless crickets and beetles. This combination of features provides the open spaces most suitable for tuataras to forage in and also favours a very substantial production of the food on which they live. Many of the burrows constructed by the petrels are used by tuataras, not only during the months that the birds are at sea but sometimes in company with birds incubating eggs or rearing chicks. The occurrence of a tuatara and petrel in the same burrow has often been described, but the reasons for the association are obscure, as tuataras can, and frequently do, construct burrows quite independently of the birds.

Offshore islands that lack some element of this combination of features do not have tuatara colonies now, nor is there any evidence of their past presence. Sub-fossil remains from mainland deposits in some limestone areas, early Maori kitchen middens, and peat bogs show that the tuatara was originally distributed through a number

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of localities ranging from North Cape at the tip of the North Island of New Zealand to Bluff near the southernmost point of the South Island. There is as yet no evidence of the occurrence of the animal at Stewart Island in the extreme south. The ecological conditions of the mainland sites at the time of the occurrence of the tuatara are not clear, but it is interesting to find that, in at least some instances, petrel bones occur in association with tuatara bones. It seems probable that a gradual change in climatic conditions, plant cover, and perhaps bird associations before and during early Maori times, was responsible for the extinction of the tuatara on the mainland, and there is no firm evidence that the disappearance was caused by introduced animals or European settlement. Despite several reports of tuataras being present on the mainland in European times, it has not been possible to rule out the probability that they were brought over from neighbouring islands and then were released or escaped on the mainland. Intensive investigation has not verified the presence of any self-supporting mainland colonies in European times, and although food remains show that the earliest Maoris sometimes ate tuataras, there is no real evidence that this was a major factor in exterminating them on the mainland.

MORPHOLOGY

In appearance, the tuatara (figure 10) shows many superficial resemblances to certain lizards, and it was indeed temporarily classified as a member of the agamid group—the Old World equivalent of iguanas. The shape of head, the body proportions, and the well-developed crest (figure 11) on the back of head and along the dorsal mid-line are similar to those of a number of true lizards, and the separation of the tuataras from these is based mainly on skeletal features. There is a marked sexual dimorphism (figure 9); the male reaches a maximum weight exceeding 1 kg and a snout–vent length of up to 290 mm, whereas the female very rarely exceeds 500 g in weight, with a snout–vent length of 240 mm. In both sexes the tail, if complete, slightly exceeds the snout–vent length, but in natural conditions most animals show loss of some portion of the tail. Reports of animals considerably exceeding 600 mm in total length have not been confirmed by the recent work.

The adult male tuatara has a massive head with a considerable development of the jowl (figures 12 and 13) and frequently shows a black patch in the fold of skin below the ear region. The female has a relatively broader abdomen, and the more delicately constructed head is rarely carried so high as that of the male. In most museum collections, males markedly predominate, probably because of their greater size and more spectacular appearance, which would appeal to many field collectors, rather than because of any great numerical difference between the sexes. Males are seen much more frequently basking at burrow mouths in daytime than are females, and even at night, males tend to be much more frequently encountered under many weather conditions. It is only during misty rain after a prolonged dry period that one encounters females as abundantly as males, suggesting that the sex ratio may really be approximately equal, but that the females are much more secretive in their habits.

BREEDING

During a typical year on Stephens Island the animals undergo a partial hibernation during the winter months; but on sunny days some will be seen occasionally at the mouths of burrows with heads and shoulders visible, and on an unusually mild night a few will be found foraging. During August, most of the population appear to become fully alert, emerging from their burrows shortly after dusk and either sitting motionless in the open, or actively searching for ground insects until the early light of dawn, when they return underground. Egg-laying (figure 2) has been noted through October, November, and December. The female excavates a special nest-depression, approximately 130 mm deep in friable, well-drained soil, and lays from eight (possibly less) to fifteen eggs in a clutch. The eggs (figure 3) have a flexible parchment-like shell, are blunt at both ends, are about 30 mm in length, and weigh 4–6 g. In two cases in the field, we found that all the eggs were laid in a single night, but specimens in captivity have laid eggs at intervals over several days. After egg-laying, the nest is filled in by the female and no further attention is paid to it by either sex. Mating has been observed in the field on only one occasion, in the month of January, and twice in captive specimens in the same month. This rather slender evidence would suggest that fertilization occurs about ten months before the eggs are laid, but the records of turtles and snakes that have laid fertile eggs after much longer periods in solitary confinement show that a number of reptiles can store viable sperm for very long periods [5, 6].

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FIGURE 1 – *Upper slopes of Stephens Island, showing herbaceous cover, lighthouse, and (top right) tiny remnant of original coastal forest cover.*

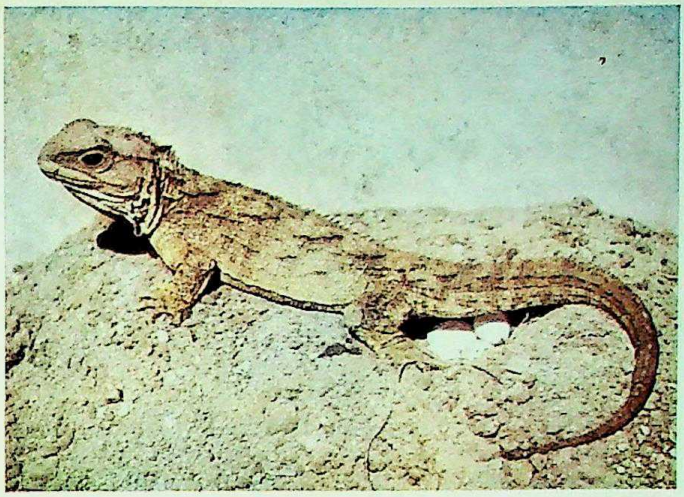

FIGURE 2 – *Captive tuatara laying eggs.*

FIGURE 3 – *Egg of the tuatara. The scale is in centimetres.*

FIGURE 4 – *Close-up of head of baby tuatara, showing egg-breaker.*

FIGURE 5 – *Emergence of young from egg that was artificially slit open.*

FIGURE 6 – *Nest seen from above, showing shrunken egg shells and hatching tuataras.*

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FIGURE 7 – *Newly hatched young, showing concealing coloration. The coin is 2·9 cm in diameter.*

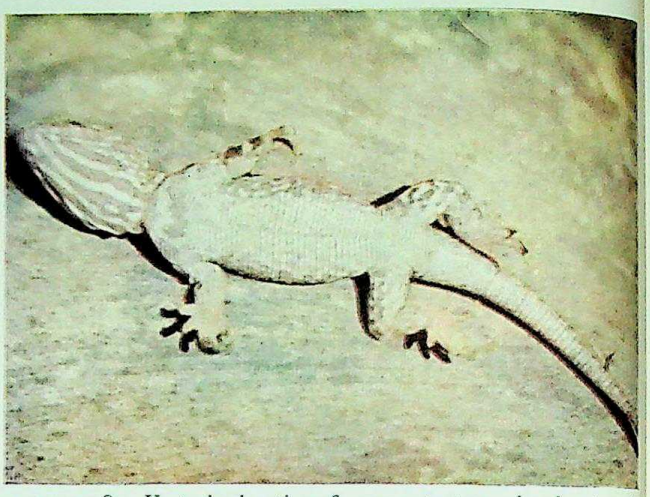

FIGURE 8 – *Ventral coloration of a young tuatara, showing prominent stripes in the throat. These are typical of young animals but fade as they become mature.*

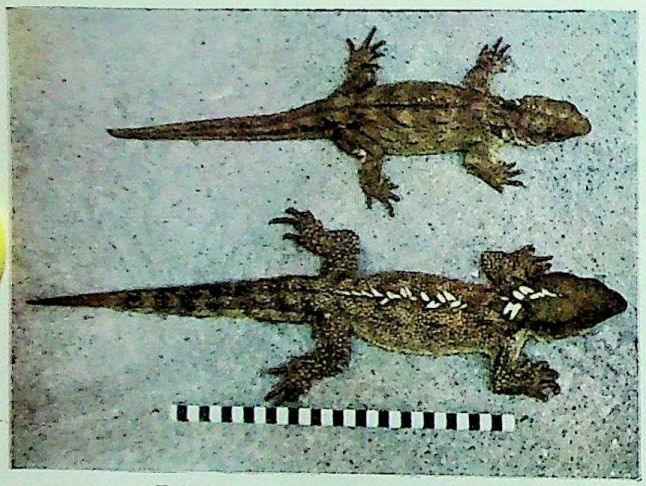

FIGURE 9 – *Fully grown male (bottom) and female tuataras, showing the greater crest-development and total size of the male, and the more delicate head and broader abdomen of the female. The scale is in centimetres.*

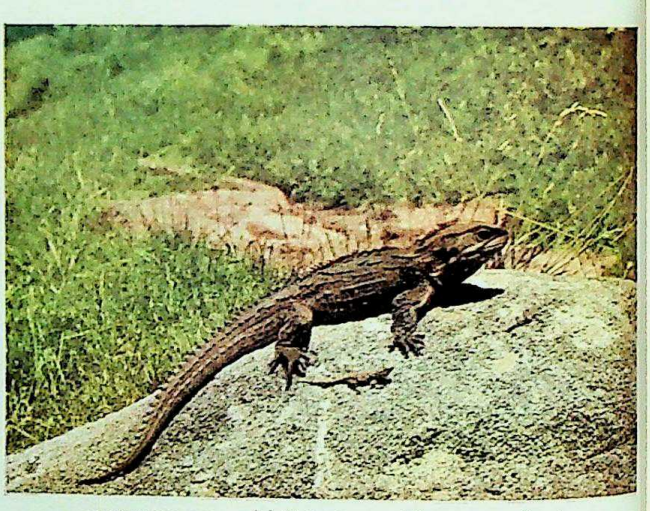

FIGURE 10 – *Adult tuatara with young animal.*

FIGURE 11 – *Tuatara moving among the leaf litter of the open forest floor, showing full raising of the crest.*

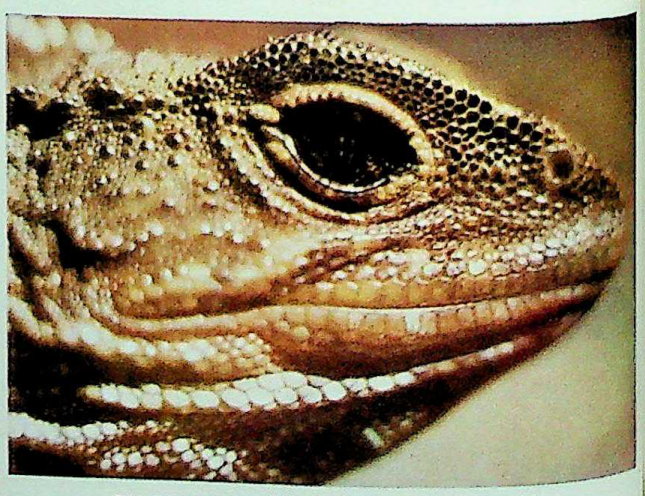

FIGURE 12 – *Head of adult seen from side.*

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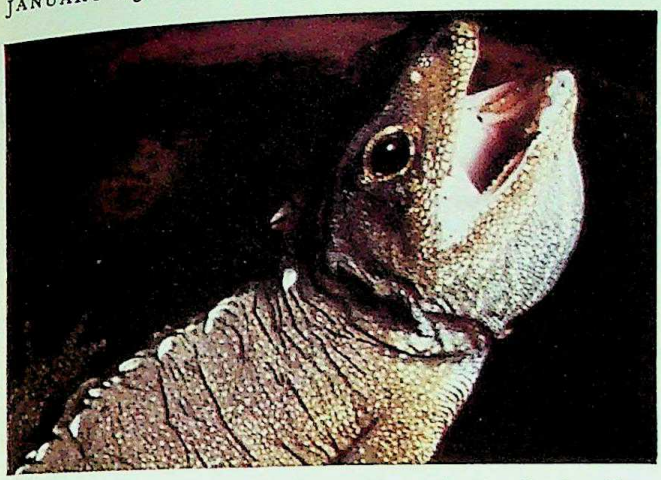

FIGURE 13 – *Fully grown male, croaking viciously after handling, showing the massively developed jowl.*

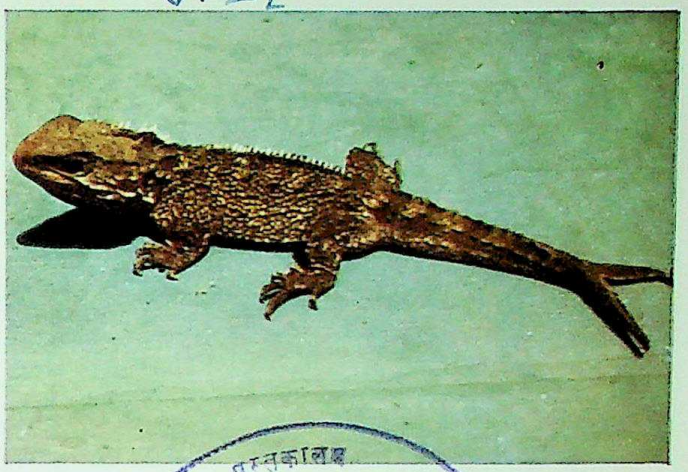

FIGURE 14 – *Forked tail resulting from regeneration in a half-grown specimen.*

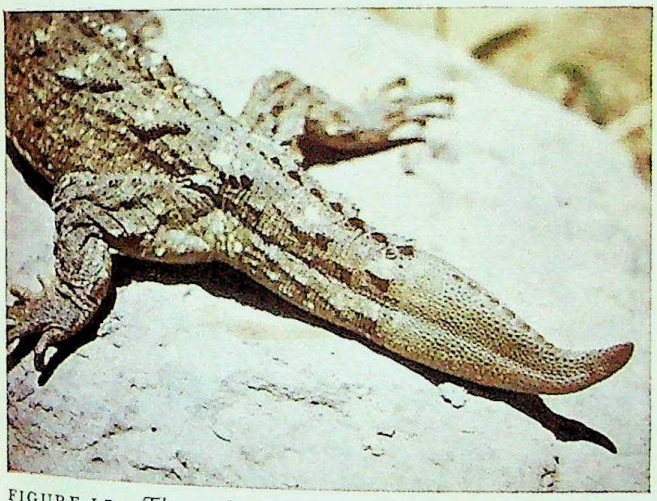

FIGURE 15 – *The marked difference in colour and texture shown between the original portion of the tail and the part that has regenerated.*

FIGURE 16 – *The appearance of the end of a tuatara's tail that has freshly lost the terminal portion.*

FIGURE 17 – *Another example of a forked tail in an adult specimen.*

FIGURE 18 – *Old male, showing stump of the left foreleg completely healed after loss of the limb high up in the humerus.*

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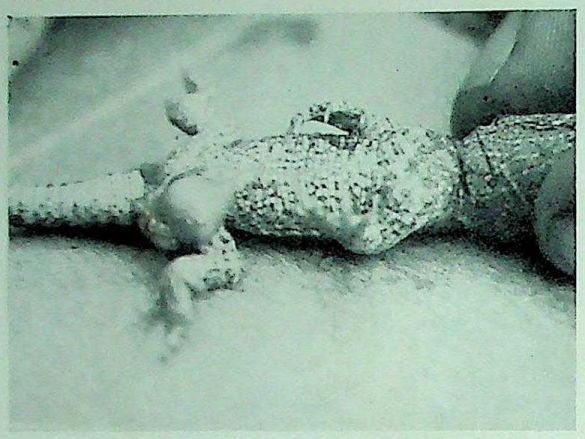

FIGURE 19 – *Ventral view of newly hatched young, showing incompletely absorbed yolk sac still attached.*

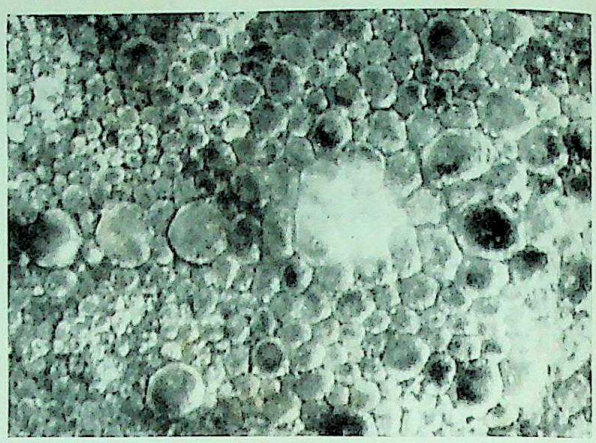

FIGURE 20 – *Pineal eye surrounded by petal-like scales in newly hatched young.*

HATCHING

Hatching has been described as occurring about fifteen months after the eggs are laid [7], and this was confirmed from a clutch kept recently under observation. Part of this lengthy incubation is taken up by a period of quiescence during the winter months, but although occasional quiescence, and consequent retarded development-rate, is known to occur in some other reptiles in special conditions [4, 5], there are no other recorded examples among reptiles of incubation exceeding nine months; the great majority require three to four months or less. The young from two tuatara eggs maintained in the same jar emerged two weeks apart, but in some nests noted in the field several young emerged during one day.

A week or two before the young emerge, the eggs absorb moisture and become slightly swollen. The parchment-like shell is slit longitudinally by a horny egg-breaker (differing from the true egg-tooth of all lizards) on the tip of the nose (figure 4). After this process, the egg and emerging young resemble the example shown in figure 5.

There was a flow of fluid into the soil, from the two eggs observed under soil in a jar, after the egg-breaker had slit the shell. It was evident that this allowed the soil to be more easily compacted by the head movements of the young tuatara as it forced its way through the slit in the egg membrane. Convulsive movements of the head and then of the body, continued at sporadic intervals through eight hours, enabled the young animal to work its way through some five inches of earth to reach the surface. On emergence (figure 6) the shrunken, but still moist, remnants of the mid-ventral yolk sac were present and persisted for a few days before drying and falling off (figure 19). The egg-breaker seldom persists for more than two weeks. The young animals (figures 7 and 8) show very slight traces of a crest, are earth-coloured, and are just as active in their movements as geckos and skinks, which I have observed under similar conditions. They are capable of constructing small burrows for themselves within the first few days, but frequently make use of available cover by secreting themselves in spaces under boulders or fallen logs. The protective coloration (figure 7) persists at least six years, and during that time crest development becomes apparent.

When the young are newly hatched, the pineal eye (figure 20) is strikingly prominent as a median uncovered spot on the top of the head, surrounded by scales radiating like the petals of a daisy. The uncovered spot becomes concealed by opaque scales in about six months. A number of attempts to elicit responses from very young animals by concentrating beams of varying wavelengths on to the pineal eye while the true eyes were covered have failed to produce any consistent response. It is of interest that recent experiments to test the pineal eye as a possible thermo-receptor likewise gave inconclusive results [8]. Although some literature on the tuatara has placed a great deal of emphasis on the well-developed nature of the so-called 'third eye', a number of lizards (and some other animals) have a comparably developed pineal eye.

GROWTH AND FEEDING HABITS

Growth of the tuatara in the field has been studied in my programme since 1950, and the 750 animals marked ranged in size from newly hatched to fully grown. More than 200 have been recovered at intervals up to nine years after marking,

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and the data so obtained have been fitted to the Bertalanffy growth formula for both length and weight increments. The result indicated a very much slower growth-rate than that so far recorded for any lizard and shows that measurable increments occur during at least fifty years and probably for a good deal longer [9].

In addition to growth-rate studies in the field, an animal hatched eleven years ago has been weighed and measured at regular intervals. During the first five years, when in New Zealand, its rate of increment was much the same as that of comparable animals recaptured in the field. Since transfer to the warmer climate of Sydney six years ago, its annual growth-rate has been greater than that shown by young of a similar size-range on Stephens Island. It has now reached a weight of 300 g from a birth weight of 4 g. Crest development and body shape suggest that the animal is a male and therefore has a potential maximum weight of about 1000 g.

Sexual maturation occurs, of course, before the animals grow to full size. The number of preserved tuataras of intermediate size available for dissection is too small to fix the minimal breeding size with confidence for either sex, but no specimens smaller than the size reached in twenty years' growth on Stephens Island have been found with mature gonads. This suggests that twenty or more years are required before the onset of sexual maturity; if this is so, the period is much longer than that established for any other reptile. Most lizards breed after three years or less, the Mississippi alligator after six years, and even the long-lived tortoises and turtles range, according to species, from after two or three years to a recorded maximum of twelve to sixteen years [4, 5].

So far there are insufficient data to give a reliable estimate of potential longevity in the tuatara, but from its slow growth and late maturation, it seems likely that many of the large animals could well be a century or more old. If this is so, many large specimens now occurring in much modified vegetation—for example, in sheep-grazed grassland, which has replaced much of the former coastal forests on Stephens Island—have in their own lifetime passed through a very substantial change in conditions without visible detriment.

A high proportion of tuataras show loss of portions of the tail (figure 16) or other injuries to limbs (figure 18), including two cases where a limb had been lost at a level half-way up the humerus. These cases are difficult to understand in the light of the absence of any observations of fighting during field observations extending over more than ten years. Damaged tails regenerate (figures 14, 15, and 17) extremely slowly if injury occurs when the animal is sub-adult or older, but can do so rapidly in the young.

Tuataras seem to feed readily on most of the ground organisms of appropriate size in their environment. The young feed mainly on smaller animals, such as earthworms, and small insects; larger specimens feed primarily on wingless crickets, large beetles, and snails, and also on the eggs and occasionally the young of the petrels that breed on the same islands. Detection of food seems to depend mainly on sight rather than on smell or hearing, and tuataras seldom react to an object until some movement occurs. They then approach the quarry, tilt their head on one side and downwards towards the object for some seconds before lunging and picking it up by combined tongue and jaw action. The object is then chewed, sometimes for several minutes.

On most of the islands, pools and streams are absent, so that the water supply is restricted to puddles that occur after small showers; there are periods when the animal may have for many weeks to depend solely on water in the food or from dew. In view of this, and of the very long incubation period of the egg, it is not surprising that nitrogenous excretion occurs mainly in the form of semi-solid white bodies consisting almost entirely of urates, and that the volume of liquid normally expelled is small, although the animal can, when disturbed, eject substantial quantities of colourless liquid containing some urea.

TEMPERATURE RESISTANCE

Tuataras can tolerate a wide range of temperature conditions, coming out to forage in temperatures as low as 7° C, and toleration of low temperature has been claimed as further evidence for the primitiveness of the tuatara compared with modern lizards [10]. However, at least two true lizards (species of gecko) that occur on many of the same islands as the tuatara can also be encountered foraging at night in similarly low temperatures. This shows clearly that toleration of low temperatures is likely to be an adaptation to nocturnal conditions in cool temperate regions, and there is no justification for distinguishing the tuatara from all lizards on these grounds. Captive specimens in terraria heated throughout the year have had a very high mortality rate in several countries, but some specimens kept outdoors throughout the seasons in Sydney have

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withstood temperatures exceeding 40° C for some hours at a time during four summers without evidence of discomfort. However, during the writer's absence all except one of a colony of eight died when air and burrow temperatures exceeded 40° C for four consecutive days, though the deaths did not occur until the end of the fourth day.

METABOLISM AND PHYSIOLOGY

Many writers have emphasized the sluggishness of the tuatara in captivity, but most of the specimens observed in this way were probably of a great age; in natural conditions, even these can run away from a threat as rapidly as most lizards of comparable size. The very young are not noticeably less agile and alert than small lizards. The only published account of metabolism describes results [11] on what appears to have been an old captive animal; some recent tests on younger animals appear to indicate that the metabolic rate may be a good deal higher than is suggested by the earlier work.

It is clear that the long history of conservatism in structure in the tuatara played a major part in the interest aroused by this species, but it is dangerous to presume that a similar conservatism necessarily occurs in the animal's physiology and mode of life. It is certain, for example, that its Triassic ancestors could not have lived in a coastal-forest marine-bird association as the modern tuatara does. Few of the modern flowering plants, and no birds, existed, let alone the specialized varieties that form the complex pattern of life on present-day tuatara islands. The subterranean habit, and especially the association with petrels in the burrows, are obvious specializations that, combined with nocturnal life, have provided a sheltered existence that was perhaps conducive to survival in recent times, but could hardly have been unchanged since the Mesozoic. The tuatara's temperature-tolerance is similar to that of the geckos living in the same areas, despite the long independent evolutionary histories of these reptiles. While it is extremely tempting to identify features of the tuatara life-cycle as conservative relics, it is obvious that many more changes in environment and mode of life must have occurred than the conservatism in the skeleton indicates. However, the extraordinarily long growth-period, the late maturation, and the metabolic rate are of sufficient intrinsic interest to invite more study. In particular, the satisfactory agreement of growth data with the Bertalanffy formula provides on the one hand a means of testing metabolic rates at various ages against theoretical expectation, and on the other a basis for estimating mortality rates against recruitment rates.

CONSERVATION

Subjectively, it appears that the Cook Strait island populations are maintaining a satisfactory replacement rate and age distribution. But the survival of a group whose distribution has contracted from being almost world-wide in Triassic times to one populating a few small islets off New Zealand at the present day must, so far as is humanly possible, be safeguarded. Its total extinction would sever a fascinating link with the Mesozoic, and the rigid protection of the species by the New Zealand Government must be applauded by all those interested in the living relics of an ancient world.

ACKNOWLEDGMENT

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Colour television

D. GABOR

Although colour television was demonstrated in 1940, it has not yet become commercially fully successful. Many of the problems were solved by the development of a 'compatible' system of colour television that would give a black-and-white picture on ordinary sets and required no wider wavebands than are used in black-and-white transmissions. This article describes the theory of compatible colour television, and also describes the different television tubes that have been developed, including one in the form of a box only a few inches deep.

Colour television poses two distinct major problems. One is the transmission of colour information in addition to monochrome information, and accommodating this in the available wavebands. The other is the production of a reliable domestic receiver at a reasonable price.

A high-definition television picture contains about 150 000 independent 'picture points'. These must be transmitted 25–30 times per second, which means that the television signal must be capable of conveying at least 4 million independent data per second. It is shown in communication theory that this requires a waveband of at least 2 Mc/s. This is a consequence of Fourier's theorem, as may be shown by the following argument. Any signal, however complicated, can be represented as the superposition of simple sinusoidal oscillations of various frequencies. If we want to transmit 4 million data in a second, we must have 4 million simple harmonic 'Fourier components'. According to Fourier's theorem, these components are spaced by one cycle per second if we carry out the analysis once per second, and there are two components for every partial frequency, which are in the relation of sine to cosine. Hence 4 million picture points require a waveband of 2 Mc/s. In practice, the wavebands used are somewhat wider, because transmission in the minimum waveband, though technically possible, requires very complicated receivers. Because of this, and of the differences in national standards, the wavebands per television channel are 5 Mc/s in Britain, 6 Mc/s in the United States, 7 Mc/s in Europe, and 14 Mc/s in France. If one considers that these are sufficient for 550–1500 sound-broadcasting channels, it is easy to understand why television has always suffered from congestion of wavebands.

Were it not for this congestion, and also for the enormous capital invested in monochrome home receivers, colour television would be comparatively easily achieved. In fact, high-quality colour television broadcasting was demonstrated as long ago as August 1940 by P. C. Goldmark, using the network of the Columbia Broadcasting System. The receiver was a white-phosphor cathode-ray tube, with a colour sector disc rotating in front of it that changed the colours 120 times per second in synchronism with a similar disc in the transmitter. This speed was later increased to 144 times per second. There were 24 colour frames, each transmitted in 6 sectors, interlaced with other colours. A waveband of 12 Mc/s was necessary for a picture with the same definition as would have required 6 Mc/s in monochrome. This, added to the disadvantage that the large rotating disc almost doubled the size of the set, prevented the success of this system, in spite of the facts that the quality of its pictures remains unsurpassed to this day and that the receiver was cheaper than one using any other system.

The difficulty was not only that a waveband twice as wide as usual had to be provided, but that it had to be provided outside the occupied wavebands. This objection appeared to be final until 1949, when the first possibility of a compatible system emerged, that is to say, of a system which occupies only as much bandwidth as a monochrome transmission, and which moreover can

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Was born in 1900 and educated at the Technical Universities of Budapest and Berlin; he worked in the German electrical industry until 1933, and in the British electrical industry from 1934 to 1948. He became Mullard Reader in Electronics at the Imperial College of Science and Technology in 1949, and has held the chair of Applied Electron Physics since its creation in 1958. He was elected Fellow of the Royal Society in 1956.

His work has been in electrical and electronic engineering, and in applied physics, with particular interests in the development of high-speed oscillography, gas discharges, physical optics, communication theory, electronic devices, and 'intelligent' machines.

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be accommodated in an already occupied waveband, because the colour transmission can be received in monochrome on ordinary sets. This is indeed the only practicable approach, if one considers the colossal capital invested in television sets. The new service must be gradually introduced, and the customers left free to choose it or not. Unfortunately there is no compatible way of going from the British 405-line system to the European 625-line standard, or else the discussion which has gone on for many years would have come to an end long ago.

Compatible transmission solved the problem of the waveband congestion in a brilliant way that is one of the major engineering achievements of our time, but it set a tough problem to the designers of domestic receivers, one that has not found a fully satisfactory solution to this day. In order to appreciate this it will be necessary to explain first, at some length, the principles of transmitting colour information in a restricted waveband.

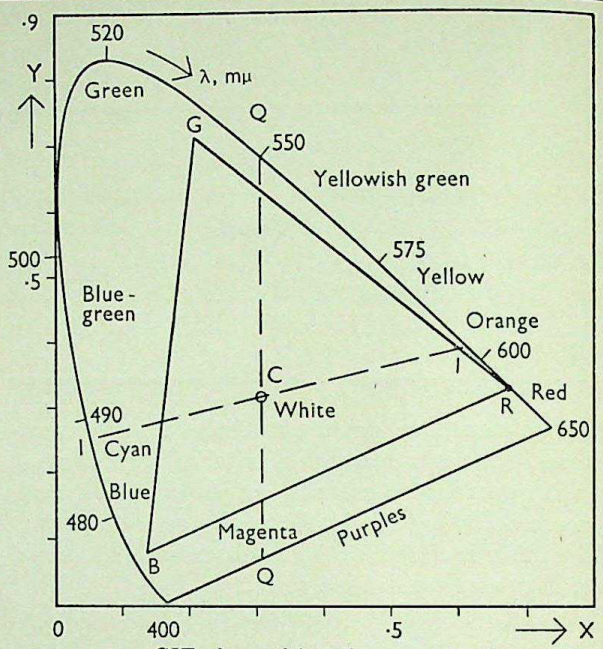

FIGURE 1 – *CIE chromaticity diagram, showing the triangle of the FCC primaries and the two NTSC colour axes I, Q.*

THEORY OF COLOUR REPRODUCTION

The science of colour vision started when Isaac Newton first decomposed white light into its monochromatic components, demonstrated that these could not be decomposed any further, and showed that their addition resulted in the original white colour. In mathematical language this means that any complex light stimulus is describable by a spectral function $E(\lambda)$, which is the energy per unit wavelength λ. But this implies that the physical stimulus has an infinity of co-ordinates, because $E(\lambda)$ can be varied freely and independently at any λ. Newton's incomparable genius immediately took a further enormous step by recognizing that the subjective light-impressions formed a manifold of only three dimensions, in which intensity is one co-ordinate, and the colours occupy two dimensions. He conceived the 'colour triangle', in which all, or, to be precise, almost all, colours are the resultants, by addition, of the three primary colours, red, green, and blue. Newton's triangle is the ancestor of the 'chrominance diagram' shown in figure 1. After Newton, Clerk Maxwell made the greatest individual contribution to this, and after him came some hundreds of patient workers. In 1931 the C.I.E. (*Commission Internationale d'Eclairage*) was formed, and the standards shown are those established in 1951.

How can one give a quantitative measure of subjective impressions, such as light and colour sensations? The answer is that the thing measured must belong to the physical, objective world, and that the subject has only to say whether different quantities give distinct sensations or not. Newton's principle is equivalent to the statement that there must exist three physical quantities, X, Y, Z, such that whichever way these are produced out of the infinite variety of the $E(\lambda)$, the subjective sensation will be the same. Faced with such a problem, the mathematician would say that the simplest way in which to produce three co-ordinates out of an infinity is by the additive hypothesis. This means that there are, for people of normal vision, three functions of wavelenths $\bar{x}(\lambda)$, $\bar{y}(\lambda)$, $\bar{z}(\lambda)$ such that all the information that the eye can receive from a surface uniformly illuminated with any colour is contained in three numbers, called stimuli:

$$X=\int E(\lambda)\bar{x}(\lambda)d\lambda \quad Y=\int E(\lambda)\bar{y}(\lambda)d\lambda$$

$$Z=\int E(\lambda)\bar{z}(\lambda)d\lambda$$

It so happens that this simplest hypothesis quite adequately represents the facts. The problem is only how to find the functions $\bar{x}$, $\bar{y}$, and $\bar{z}$. It would be a very difficult one were it not that one can divide the visual impressions into two parts, luminance and chrominance. One can fairly well compare the total brightness or luminance of even very different colours with the flicker photometer, which presents them in rapid alternation and

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allows us to match them in brightness by using a neutral filter. One can therefore imagine the luminance as a third co-ordinate at right angles to the co-ordinates xy of the chrominance diagram in figure 1. It is even better to visualize this figure as a section of a pyramid, with the apex somewhere behind the plane of the drawing, because at zero luminance there is only one colour, black.

The chrominance diagram is a two-dimensional ordering of colour sensations, with the chrominance co-ordinates

$$x = X/(X+Y+Z) \qquad y = Y/(X+Y+Z)$$

The whole diagram is defined once one has obtained the locus of the spectral colours. This is where Maxwell made his great contribution, by giving this locus the horseshoe shape. On this horseshoe, complementary colours are on straight lines passing through the standard white point C. The lengths of the radii from C are inversely proportional to the ratio in which the two complementary spectral colours must be mixed to produce white. This is a mathematical consequence of the fundamental additivity rule. Spectral colours between blue-green and yellowish greens have no spectral opposites; their complementaries are the purples. These are located on a straight line connecting the ends of the horseshoe. The fundamental principle that all possible light stimuli must be linear compositions of spectral colours implies that this line must be straight; the extreme purples are mixtures of the two extremes of the spectrum.

It can now be seen that Newton's red-green-blue colour triangle was not quite perfect: it is not possible to cover the whole area by any triangle. This is why high-quality colour prints are made with more than three colours. One can make only a good compromise, and figure 1 shows the one recommended by the F.C.C., the American Federal Communications Commission, in 1953. The three primaries, R, G, B, can be closely approximated by existing phosphors.

Such a diagram cannot exactly reflect the subjective colour sensations, because it is not possible to make a complete separation of luminance and chrominance. It is well known that the colour brown exists only at low levels, that a faintly illuminated white sheet appears bluish, and that at high intensities even the pure spectral colours, with the exception of red, go over into white. No simple additive theory can account for these phenomena. Erwin Schrödinger in 1921 proposed an ingenious frame for a more complete representation, using a non-euclidian geometry in the three-dimensional space of visual sensations, but this has not yet been completely worked out. It is, however, irrelevant from the point of view of television transmissions, because the three stimuli X, Y, Z allow us to reproduce closely enough the same sensations in the viewer of the transmitted picture as those from seeing the original. If he would have seen brown, he will see brown, though according to the chrominance diagram he ought to have seen orange-purple.

COMPATIBLE TELEVISION SYSTEMS

This was the state of the science of colour vision when the N.T.S.C., the American National Television System Committee, started its work in 1950. More exactly, this was the state of the science of colour vision as applied to large illuminated areas. It had to be supplemented by the observation that the human eye did not ask for high definition in colour. If one takes a high-definition black-and-white transparency and colours it by projecting on top of it three colour-separation pictures, the eye will see a sharp, satisfactory picture even if the colour slides are badly defocused.

Careful tests have shown that a coloured picture appears acceptably sharp if it is built up of a sharp black and white picture on which is superposed a cyan-orange picture with only one-sixth of the definition, and a green-magenta picture with only one-sixteenth—not one-third and one-eighth as is often stated, with an error of a factor of two, which will be explained later.[1] Thus only 23 per cent more information is needed to turn a monochrome picture into a coloured picture; that is to say, it could be accommodated in a waveband extended by 23 per cent.

But in fact no extension is needed at all. P. Mertz and F. Gray showed in a classical study in 1934 that an ordinary television waveband is less than half filled. As shown in figure 2, the signal is concentrated in rather narrow bands at multiples of the line frequency. This is easily understood if one considers that vertical lines play a great part in the visual universe; horizontal lines in a television scan will therefore often be more or less repetitions of one another. If they were exact repetitions, the signal would be concentrated in sharp lines, in accordance with Fourier's theorem that any periodically repeated

[1] Definition is to be understood for these purposes in such a way that a reduction of definition to one-sixth of its previous value means that a line 0·1 mm in breadth in the original is increased to 0·6 mm.

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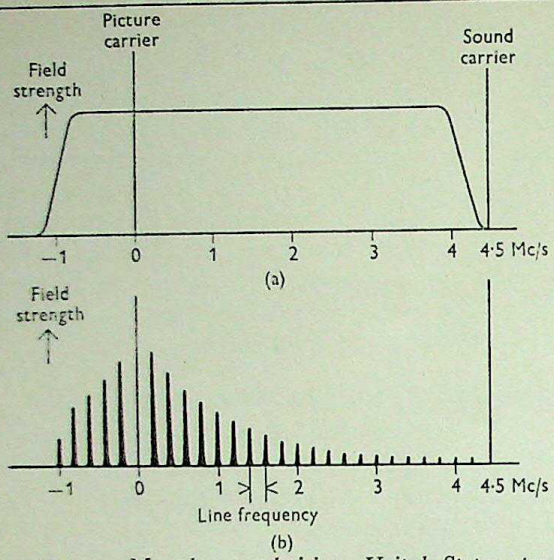

FIGURE 2 – *Monochrome television, United States standards.* (a) *Channel profile,* (b) *typical utilization.*

signal has components at multiples of the repetition frequency only. Statistics of actual pictures show that the gaps are wide enough to accommodate a second television channel.

In the NTSC system, as accepted in the United States at the end of 1953 (figure 3), the luminance is transmitted in the ordinary way and can be received by any black-and-white set. It has a channel about 4·25 Mc/s wide. Only the single sideband provides extra information; the vestigial negative sideband, extending to −1·25 Mc/s, simply repeats a part of the information, and is left only for the convenience of television-set makers. The chrominance information is understood to add nothing to the luminance. The excursions from the white point C, which add colour to the picture, are made at constant luminance, that is to say, the white component is decreased as the excursion increases. A chrominance carrier is placed at 3·579545 Mc/s from the luminance carrier, which is 455 times one-half of the line scanning frequency. This factor, 455, is $5 \times 7 \times 13$, which means that the chrominance carrier can be produced from the luminance carrier by a series of frequency multiplications, or that, alternatively, the line frequency can be derived from this difference by a sequence of reasonably simple frequency-dividers. This choice ensures that an ordinary monochrome receiver will take almost no notice of the colour transmission. All the colour sidebands will now take their place at full multiple spacings from the chrominance carrier, right in the middle, between the bandlets of luminance information.

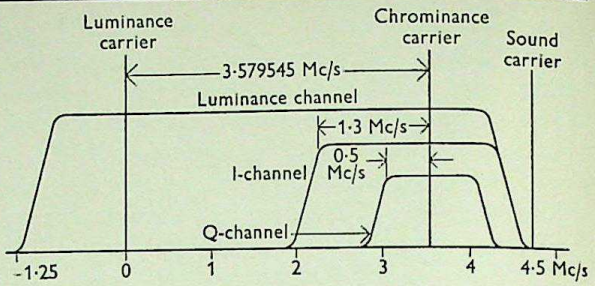

FIGURE 3 – *American (NTSC) colour television standards.*

The NTSC system has a further ingenious feature. The chrominance channel carries two independent messages, the I (cyan-orange) and the Q (green-magenta) information, with transmission bands of 1·3 and 0·5 Mc/s. These are distinguished by their phases, because they are in quadrature to one another, in the relation of sine and cosine. Figure 4 illustrates the typical utilization of the waveband, taking account of the phases. This figure makes it clear that though, for instance, the I-channel has a single sideband of 1·3 Mc/s, this is not equivalent to one-third of the luminance channel of 4 Mc/s but only to one-sixth of it, because the phase of the chrominance channel is fixed, and therefore, unlike that of the luminance channel, not available for transmitting information. Figure 5 illustrates how the I and Q signals, mixed in the transmission, can be separated in the receiver by phase-sensitive demodulation.

The NTSC recommendations, which became United States standards in 1953, represent probably the most ingenious committee work on record. They completely solved the problem of compatible television—but at a cost. Waveband saving can never be achieved except at the price

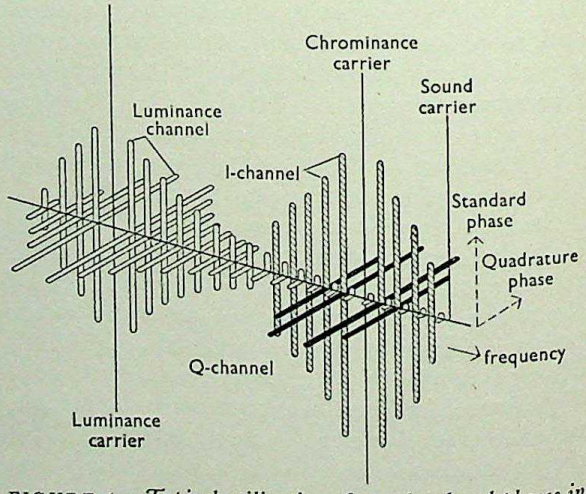

FIGURE 4 – *Typical utilization of waveband and phases in the NTSC system.*

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of complications in the receiver, with attendant dangers of malfunctioning, and these were so serious that colour television has not become a popular success in the nine years since 1953.

In the NTSC system, or in any other compatible system,[1] the colour signals are separated by frequency or phase, but not in time. This automatically excludes the field-sequential method used in Goldmark's early system of rotating filters. The colours must be produced either simultaneously or in very rapid succession, otherwise one runs into the problem of storing a great amount of information. Only electronic methods are capable of simultaneous or very rapidly alternating colour reproduction. Before we discuss the most versatile of electronic instruments, the cathode-ray tube, we must mention that frequently recurring pipe-dream, the solid-state television screen. Crystals and crystal powders that emit light when current is passed through them do exist. One can construct a panel of such electroluminescent substances, with a system of parallel conductors at the back and another system of transparent parallel conductors at right angles to the first at the front, that will light up only at the point where two energized conductors cross. This can therefore be scanned in the same way as a television picture. The difficulty is that such panels can give the light intensities required in a television picture only when they are constantly excited, while in a television scan each point is energized for less than 1/100 000 of the total time. There is not the slightest hope that electroluminescent substances could ever yield 100 000 times higher brilliancies than at present, although the cathode-ray tube can easily provide this brilliance. Hence the only possibility appears to be to give each picture point its own energy supply, set to a certain intensity by the television signal and reset when the next frame comes along. The problem of using more than 100 000 miniature amplifiers is one that few inventors would care to contemplate. There is

[1] One alternative to the NTSC system is the French SECAM system, short for *Système Séquentiel à Mémoire*, invented by Henri de France and developed by the *Compagnie Française de Télévision*. In this system, only one chrominance component is transmitted in any one line, the I- and the Q-components alternating in successive lines, but the component which is not transmitted is repeated from the previous line by means of a memory system consisting of a quartz delay line. The chrominance information is not frequency-interlaced, but is transmitted by frequency modulation. Another interesting alternative was proposed by J.-A. Boutry, of Paris.

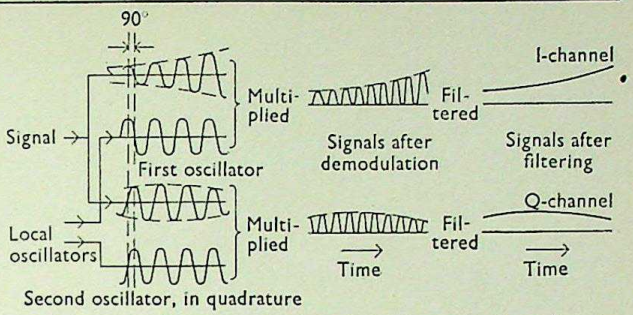

FIGURE 5 – *Phase-sensitive demodulation of colour signals. For simplicity, the signals are shown separated before demodulation. In reality, they are separated only by the demodulation, because each signal gives a non-zero modulation product only with its own oscillator.*

therefore no reason to believe that the solid-state screen will become a serious competitor to the cathode-ray tube.

ELECTRONIC COLOUR REPRODUCTION

It is known from colour photography that there are two alternative ways of colour reproduction, the additive and the subtractive, and that the second has proved far superior. Unfortunately, only the additive method can be used in cathode-ray tubes. The only successful cathode-ray device which does produce the equivalent of a transparency is the ingenious Eidophor of the late F. Fischer of Zürich, which writes ripples on a thin oil film with the charges of an electron beam. This is the best large-screen television projection device, but it is far too expensive for home receivers.

A very obvious method of producing additive colour is to take three cathode-ray tubes showing different colours, and to project the pictures on top of one another. Unfortunately this method fails, not only because three tubes are rather expensive and it is difficult to bring them to full registration, but because so much light is wasted in projection that the high brilliancies to which the public are used could be realized only at a prohibitive cost.

There are two basic methods (figure 6) for producing additive colour in one tube. The first method uses geometrical selection. In this, phosphor patches or lines of different luminescent colour are arranged side by side, and are selected by the electron beam. One may ask why one does not rather use a white phosphor with patches or lines of colour filters. The reason is that no suitable inorganic dyes exist and that organic dyes cannot stand the temperatures used in the manufacture of cathode-ray tubes. All existing television tubes are variations of the geometrical colour-selecting principle.

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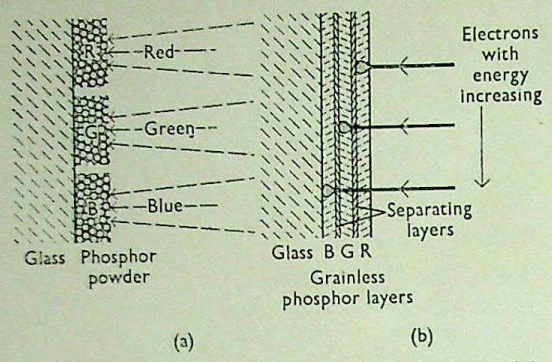

FIGURE 6 – *Colour control by geometrical selection and by penetration. The onion-shaped regions illustrate the volume into which an electron is scattered and in which it loses most of its energy.*

In the second method, three layers of phosphors with different colours are arranged on top of one another, and the selection is done by electrons of different velocity. This method has often been proposed, but it remained a dream until C. Feldman showed in 1957 how grainless phosphor screens with thicknesses of the order of a micron could be produced by evaporation, followed by baking. He showed also that most of the energy of the electrons is given up at a depth so well defined that a high degree of colour purity can be obtained. There exist now excellent tubes that 'write' in two or even three colours. However, the application of the layer screen to television tubes was not successful, because it is extremely difficult to obtain good colour registration with electrons of widely different energies.

If one uses the principle of geometrical selection, the point of arrival of the electrons must be regulated with an accuracy of about 0·25 mm, because if the patches or lines are to be invisible, a triplet cannot be larger than about 1 mm. It is impossible to regulate the arrival with this accuracy by aiming the electrons from a gun 25 cm or more away from the screen. Three methods are possible. One involves selecting the electrons by their direction. This is the method adopted in the shadowmask tubes (figure 7). The second involves distinguishing them by their time of arrival, in which case the screen must have different admission properties at different times, that is, the screen itself must contain some modulating device. In the third method, the screen sends back a signal that reports which colour is excited, so that the position can be corrected.

The only three-colour tube which has so far been made commercially in large numbers is the shadowmask tube of the Radio Corporation of America. It contains three electron-guns that are aimed at the centre of the screen. The three beams are deflected in such a way that they always converge on one spot of the screen, at angles of 6° to one another. About 12 mm in front of the phosphor screen there is a sieve-like, curved metal plate, perforated with about a third of a million small holes, the shadowmask. On the screen, behind each hole, there are three phosphor dots, so arranged that each is excited by one only of the three beams penetrating the shadowmask. In all there are about a million phosphor dots. The manufacture of this composite screen to the required precision is a highly ingenious and very delicate process. Each screen is made with its own shadowmask, used as a photographic mask. An optical system that imitates the electron optics in the finest detail is placed in the tube. Three photographs are taken in succession on a 'resist' which becomes sticky when illuminated; the light used is arranged as if it were coming from one of the three electron-guns. A colour phosphor is dusted on and is blown off the areas where the resist has not become sticky. New resist is coated on, and the operation is repeated until the dots of all three colours are in position; the shadowmask must be removed each time and put back again into exactly the same place. In all, 25 operations are needed to complete the shadowmask, and errors can be tolerated in only a very few of the million colour dots.

Another difficult operation begins after the tube is completed. However accurate the manufacture, every tube has to be adjusted by highly trained personnel. A number of small magnets are attached to a belt on the neck of the tube and adjusted until the beams converge in the centre and also in the four corners of the picture. The adjustment may have to be partly repeated *in situ*

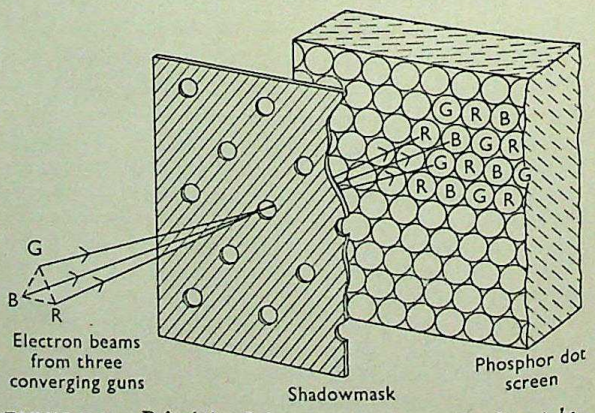

FIGURE 7 – *Principle of three-gun shadowmask colour-tube.*

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after installation at the customer's house to allow for the earth's magnetic field, which, in spite of very careful shielding, can have an appreciable disturbing influence on the colours.

One might wish that such a marvel of engineering ingenuity and boldness of enterprise had been rewarded with commercial success. Instead there were six lean years, in which the number of colour sets sold was about 1 per cent of the number of monochrome sets, and the losses in colour television were perhaps greater than in any private enterprise in the fifties. Now at last there appears to be a turn for the better. The first year to show a profit was 1960, with colour-television set sales amounting to 200 million dollars; several firms are now taking up the manufacture of colour sets, and two large firms are entering the manufacture of tubes.

The second most important method of colour control is grid focusing, chiefly associated with the great experimental physicist Ernest O. Lawrence. This is a one-gun tube (figure 8); the colours are produced not simultaneously, as in the shadowmask tube, but in succession. The screen carries a system of vertical strips of colour phosphors, of about 1 mm width per triplet. A few millimetres in front of the screen there is a double array of vertical metal wires, interlaced, and in accurate register with the phosphor strips. If the two arrays are both held at a definite potential lower than the screen potential, every pair of wires forms a cylindrical electrostatic lens such that the electron beam will be concentrated on the central, green phosphor, strip. If the potential of one of the two arrays is raised and that of the other lowered by the same amount, a deflecting field will be added to the lens field, so that the beam, still well focused, will now fall on one of the side strips, say on red. Reversing the potential

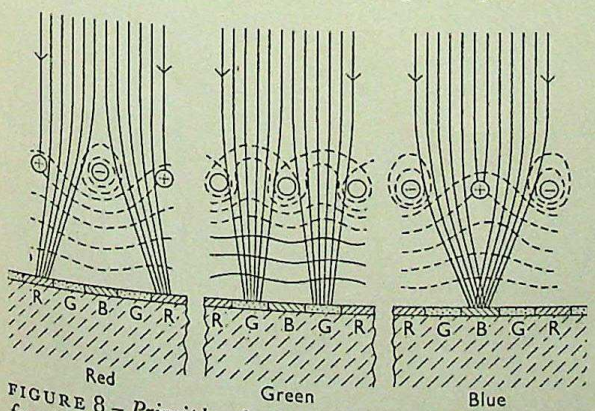

FIGURE 8 – *Principle of colour control in single-gun grid-focusing tube.*

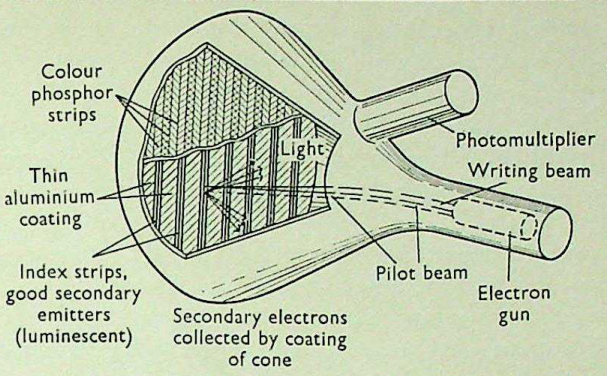

FIGURE 9 – *General scheme of a beam indexing tube. Operation by light shown in brackets.*

difference produces blue. Thus the potential difference between the two arrays must be rapidly oscillated with a frequency higher than the point frequency during the scanning, and the gun switched synchronously with this oscillation to the red-green-blue-green-red . . . signals. This is not as straightforward as deriving the signals for the three guns, as in the 'simultaneous' RCA tube, but the electronic engineers have found good solutions of the problem.

Like the RCA shadowmask tube, the Lawrence tube, which has been developed by Chromatron Ltd after many years' work, is a masterpiece of manufacturing ingenuity. It has now been developed with a cylindrical screen with a horizontal deflection angle of 110°, and hence it can be made as short as a monochrome tube. The RCA tube, with a horizontal deflection angle of 70°, is considerably longer.

In the third type of colour tube, accurate positioning of the cathode-ray spot on the phosphor strip is achieved by deriving a signal from the screen and correcting the position accordingly (figure 9). These are called indexing tubes, and their prototype is the 'apple' tube developed by the Philco Corporation. In this tube, the colour strips are vertical, and, as usual, coated with a thin layer of aluminium. At the back of this layer, indexing strips are arranged, also vertical, with a secondary-electron emission higher than that of the aluminium backing. The strips are made, for instance, of gold or of magnesium oxide. Every time the writing beam passes an indexing strip, secondary electrons are released, which are collected, and by comparison of their phase with the phase of the colour signal, small corrections are made in the line deflecting voltage.

In reality, the tube is more complicated than might appear from this description. If the colour strips are vertical, the indexing strips cannot be

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vertical, because allowance must be made for the different transit time of the secondary electrons to the electron collector from different points of the screen. This has been overcome by a carefully precalculated system of curved lines. A more serious difficulty is that the phase indication of the index strips can be swamped by the colour modulation of the writing beam; this can cause not only errors, but permanent oscillations in the feedback system. This has been overcome in the Philco tube by using a separate pilot beam, modulated with a high frequency, so that the index information can be separated out. A disadvantage common to all beam-indexing tubes is that the writing beam must be finer than a colour strip, but this has been overcome by special gun-designs. Various attempts have been made also with beam index lines with horizontal colour strips, but without great success.

The colour-tube work so far reviewed was all done in the United States. In Britain the television tube manufacturers excusably held back, waiting for the outcome of the American experiment. Nevertheless a certain amount of research was done. The Sylvania-Thorn Colour Television Laboratories have done exploratory work on an indexing tube in which the indexing is done by light, from fluorescent strips, instead of by electrons from secondary emitters. The interesting novelty is that the feedback oscillations are avoided by using a non-integral index system, so that resonance of the index signals with the colour sequence are avoided. This makes it possible to dispense with the pilot beam.

The greatest independent enterprise in this country was undertaken by the Mullard Research Laboratories, who have recently reported on a new colour display system centred round the 'banana' tube (figure 10). This is a straight cylindrical tube of about 10 cm diameter, as long as the horizontal television scan. It contains only three phosphor strips, of the three fundamental colours, which are excited alternatingly by the beam from a single electron gun. The line scan is done electronically, but the vertical scan is mechanical. A drum containing three straight glass rods rotates round the banana tube. Every one of these acts as a short-focus cylindrical lens with wide aperture, so that a reduced image of the three phosphor strips would appear, to observers placed within a fairly large solid angle, to rotate with the glass rod. The picture thus obtained would be vertically compressed and unevenly scanned. It is vertically magnified and made linear by a hyperbolic mirror fixed to the lid of the receiver.

The basic idea of this system was conceived by Nigel Calder, and a team of more than twenty workers was engaged on it for years. They have

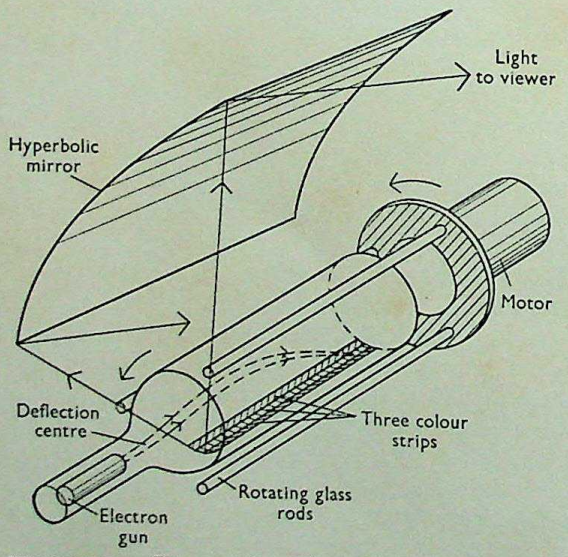

FIGURE 10 – *Banana-tube colour display system. The curved electron trajectories are produced by a permanent magnetic yoke (not shown). The rotating glass rods are shown in the position at which one has just written the bottom line of the last field, and another rod is taking over, to write the first line of the next field at the top.*

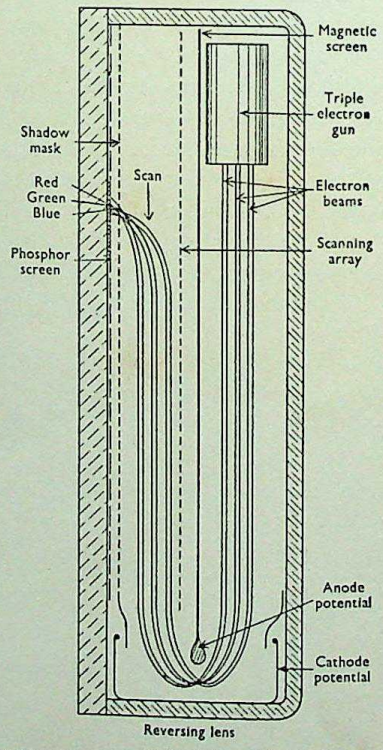

FIGURE 11 – *Principle of flat colour-tube.*

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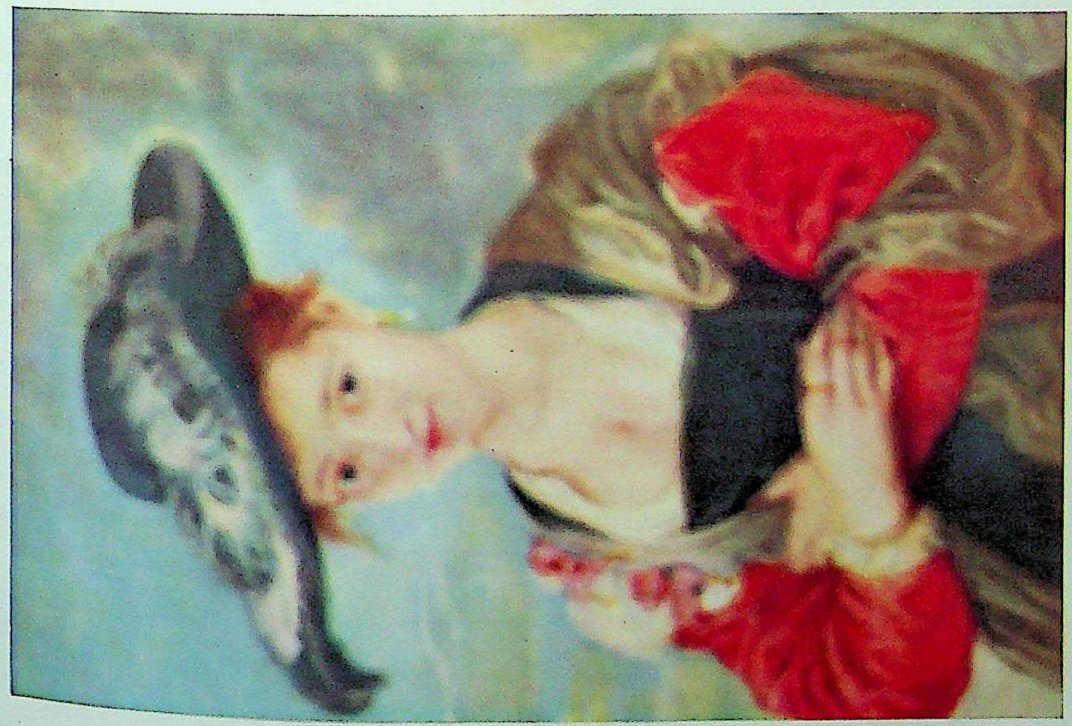

(a)

(b)

FIGURE 12—(a) *Three-colour print, strongly unfocused.* (b) *The same, overprinted with sharply focused black-and-white. This illustrates the principle of band-saving in colour television—often called the 'Principle of Mixed Highs'—with high-definition luminance component and low-definition chrominance. It is not possible to do this principle full justice by colour prints, which use a subtractive method, so that black must be added: in colour television, white is added to the chrominance background. The picture is Rubens's* Le Chapeau de Paille. *(Reproduced by courtesy of The Trustees of the National Gallery, London.)*

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recently given an excellent demonstration at the Institution of Electrical Engineers. The Mullard Research Laboratories do not claim that their system has any great advantage over others, and are very reticent about its commercial prospects.

A third line of work was pursued for several years and is still being pursued by the author at Imperial College, London. This work is directed towards a flat television-tube, in the shape of a box only a few inches deep. It has a rather intricate and novel electron-optical system which cannot be explained in full detail here. Only the essentials for colour production are shown in figure 11, which is a vertical section of the tube, at right angles to the screen. The three colour beams are launched vertically downwards into the space between the phosphor screen and a device called the scanning array. This is a system of horizontal, individually insulated, metal tapes. They receive electrical charges from the writing beam itself, which at the end of every line touches the end of one or a few conductors of the scanning array and gives them a negative charge. This makes the conductors above the beam more negative, so that an electric wave is generated, and this gradually sweeps the beams downwards. At the end of each vertical scan the whole negative charge of the array is discharged, again by electron bombardment, but this time under conditions such that the secondary electrons, which outnumber the primaries, can escape from the conductors. These therefore re-acquire a high positive potential, and the vertical scan can start again from the top of the picture.

From the point of view of colour production, the interesting feature is that, at the line scanned at any moment, the beams thrown to the phosphor screen are focused at the same time. That is to say, the convergence of three beams to one point, which in other three-gun tubes has to be effected by very delicate means, is automatically ensured here. Moreover, the convergence angle of the three beams, which is a few degrees in conventional tubes, can be here as much as 20°. This makes it possible to put the shadowmask half a millimetre from the phosphor screen, instead of 12 mm. This in turn makes it possible to stick the shadowmask solidly to the phosphor screen by means of narrow vertical ribs, invisible to the viewer, so that the tubes are much easier to make.

Though many years have been spent on this device, with some able collaborators, and although the advantages mentioned above have been experimentally demonstrated, still more remains to be done to make the flat colour-tube a commercial proposition.

It is very likely that development of this tube, as well as others, would have advanced further had it not been for the wave of pessimism which swept the television industry after the first unsuccessful years of colour television in the United States. The fact is that the United States public did not take to it with the enthusiasm one might have expected. The high price in itself is no explanation; 500 dollars for a colour set is not a larger slice of the United States worker's pay-packet than the £50–£80 that the British working man is ready to pay for his black-and-white receiver. The explanation may have been the rumours of unreliability. It may have been the insufficient, to put it mildly, enthusiasm of the radio dealers. This is certainly a part of the explanation. But the main reason was probably that the colour pictures received in the homes were simply not quite good enough, and the extra enjoyment of a few hours per week of colour transmissions was just not enough to break down the customer resistance. Outside the United States, Japan is at present the only country in which a regular colour television service has started. This was in 1960, using the NTSC system; the number of colour sets in private hands is very small, because of their high price.

There are now signs that the colour television sales in the United States are beginning to increase more rapidly. It is very probable that relatively small technical improvements, combined with somewhat lower prices and a determined expansion of colour broadcasts, will be sufficient to start a steep rise. Assuming peaceful development and reasonable prosperity, colour television is likely to become a popular success in the United States before the middle of the sixties, and in Europe towards their end.

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The non-protein amino acids of plants

L. FOWDEN

Unlike animals, plants must synthesize all the amino acids necessary for the formation of protein. In addition, however, they synthesize at least sixty amino acids which, so far as is known, are not incorporated into protein. Some of these amino acids are found in only a very few species, and many have unusual structural features not found in other natural products. The speed and ease with which amino acids can be identified by chromatography has greatly extended our knowledge of these unfamiliar substances in recent years.

Although animals are dependent upon a dietary supply of certain 'essential' amino acids, green plants and some micro-organisms are able to synthesize all the amino acids they require for the formation of protein. In consequence, plants have frequently been used in studies of intermediary nitrogen metabolism, especially in recent years, when refined chromatographic methods have made amino-acid analysis much easier. An unexpected feature of these investigations has been the discovery of a large group of amino acids that are not normally components of protein, and that are certainly not artefacts produced during the isolation procedure. Certain novel chemical structures, not seen within the protein amino acids, are encountered in some members of the new group.

It is natural to ask what function these compounds serve in plant metabolism and growth. Are they involved in the synthesis and degradation of protein amino acids? Are they important in nitrogen assimilation, storage, or transport? Have they any regulatory or controlling action upon metabolic rates and hence indirectly on growth? Or are they mere 'accidents' or by-products of an aberrant metabolism, perhaps even evolutionary misfits not yet discarded? Provisional answers can be given for some of the compounds, but many of the acids seem destined to remain as chemical curiosities for a long time, because it is difficult to study the metabolism of the plants in which they occur. Indeed, the degree of uncertainty that is at present associated with the function of these compounds resembles that surrounding other types of plant products, including the alkaloids, floral pigments, essential oils, and polyphenols.

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HISTORY

The task of identifying the amino acids present in plants has been a sporadic rather than a continuous process. Only a few years elapsed between Lavoisier's recognition of the elementary character of nitrogen and the first isolation of an amino acid from a plant material. The crystallization of asparagine from the juice of asparagus shoots in 1806 by L. N. Vauquelin and P. J. Robiquet [1] prompted other isolations of this amino-acid amide from plant sources during the following half-century. Before 1860, evidence for the existence of other amino acids in plants was inconclusive, and the relative ease with which asparagine could be isolated led many plant physiologists to adopt the erroneous idea that plant proteins contained only asparagine and carbohydrate, combined in different relative amounts.

A second phase of development began with the pioneering analytical studies, between 1860 and 1872, of H. Ritthausen, who established that several other amino acids were present in hydrolysates of vegetable proteins. The new isolations stimulated Ernst Schulze to start his brilliant and painstaking series of investigations of the protein metabolism of growing plants. These studies, which continued for nearly 40 years, further increased the number of recognized amino acids, and led to the general acceptance of the importance of amino acids in protein structure, paving the way for E. Fischer's peptide hypothesis. At the end of Schulze's active researches (1906), fifteen amino acids, including the amides asparagine and glutamine, had been identified as plant

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constituents. We now know that all these acids, together with five others discovered subsequently, occur in the free state, and combined in proteins, in all plants, almost without exception. Excellent and full accounts of these early investigations have been given by A. C. Chibnall [2] and D. M. Prianischnikow [3].

As there were no methods for the rapid specific recognition of amino acids during this classical period of investigation, the characterization of particular amino acids in plants depended upon the application of laborious isolation techniques. The paper-partition chromatographic technique for the separation and identification of amino acids was introduced in 1944 [4]; it provided a sensitive routine method for the study of free and combined amino acids in plants. The complete spectrum of amino acids present in a particular plant can now be ascertained in a few days, whereas the methods available to Schulze required months, or even years, of industrious labour, which, even then, gave a less comprehensive result. It is therefore not surprising that the advent of paper chromatography heralded a third phase of amino-acid discovery, in which, to date, about sixty compounds have been added to the list of substances found in plants.

The amino acids of this new group differ in two important biological features from those isolated earlier; firstly, their presence is normally restricted to relatively few plant species, and secondly, they usually exist only in the free state. They can be described loosely as non-protein amino acids. The progress made in the identification of non-protein amino acids is shown in figure 1. As yet there is no indication that the steeply rising curve has reached a maximum.

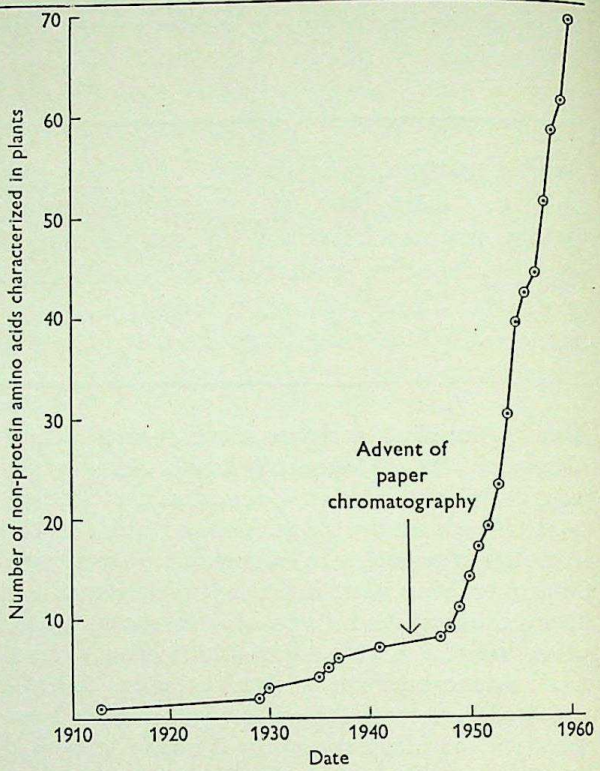

FIGURE 1 – *The effect of paper chromatography on the rate of identification of new non-protein amino acids.*

IDENTIFICATION AND ISOLATION OF NEW AMINO ACIDS

Paper chromatography provides an ideal method for surveying the amino acids present in a large number of plant extracts. The procedure is extremely sensitive (a few μg of an amino acid give a detectable spot after treatment with ninhydrin) and chromatograms such as those illustrated in figures 2a and 2b can be obtained from about 0·5 g of plant material.

Most of the protein amino acids are always present free in a plant extract, and so their ninhydrin-reacting spots give a chromatogram a definite and reproducible pattern. Occasionally, a plant extract yields a spot differing either in its position or ninhydrin colour from those of all amino acids previously recorded; if the intensity of the spot indicates that the plant contains appreciable amounts of the substance, the isolation of yet another new amino acid becomes a possibility.

A new amino acid is normally isolated by running an extract of plant material through a suitable cation-exchange resin (H^+-form), when all amino acids are absorbed; non-cationic substances may be washed completely through the column. The amino acids are displaced from the resin using dilute solutions either of ammonia or of buffers containing volatile ammonium salts. Usually, a partial separation of the amino acids is obtained if the eluate is collected in fractions [5]. Figures 2c and 2d illustrate the fractionation of the amino acids present in an extract of shoots of white bryony (*Bryonia dioica*) that contained two new substituted asparagines. The isolation of pure substances from enriched fractions requires additional separations, by either further resin-column chromatography or large-scale paper chromatography.

STRUCTURES OF THE NON-PROTEIN AMINO ACIDS

Protein amino acids all conform to the general structure, $R.CH(NH_2).COOH$. Specific properties

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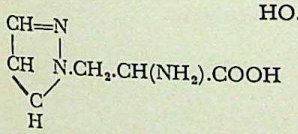
are conferred upon individual acids by variation of the radical R, which may contain acidic, basic, hydroxyl, aromatic, heterocyclic, or sulphur-containing groups. The same general formula satisfactorily describes the majority of the non-protein amino acids, although examples of non-α-amino acids are known, and include γ-aminobutyric acid, β-alanine, and β-aminoisobutyric acid. Table 1 shows a selection of structures derived for the non-protein amino acids. More comprehensive lists have been given in several reviews [6–9]. Many of them are chemically closely related to the protein amino acids, being either homologues or simple substituted derivatives, and they can be classified according to the usual scheme adopted for the protein amino acids.

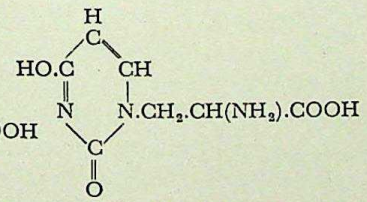
$\overset{\gamma}{C}H_2(NH_2).\overset{\beta}{C}H_2.\overset{\alpha}{C}H_2.COOH$
γ-Aminobutyric acid

$CH_2(NH_2).CH_2.COOH$
β-Alanine

$CH_2(NH_2).CH(CH_3).COOH$
β-Aminoisobutyric acid

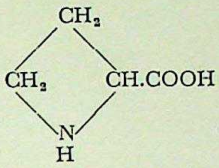
Certain distinctive structural features are also encountered. The pyrazole ring, present in β-pyrazol-1-ylalanine, is not known to occur in any other natural product, and the amino acid is one of the very few natural products that contain linked nitrogen atoms.

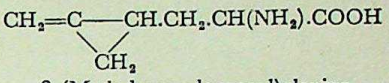

β-Pyrazol-1-ylalanine


β-Uracil-3-ylalanine

Several of the non-protein amino acids may be regarded as β-substituted alanines and in this respect may be compared aptly with such protein amino acids as histidine, phenylalanine, and tryptophan. However, in β-pyrazol-1-ylalanine and β-uracil-3-ylalanine, from seeds of water melon and *Acacia* respectively, the novel situation exists in which the alanine moiety is attached not to a carbon but to a nitrogen atom of the ring system.

Cyclopropane and azetidine structures are uncommon in natural products; the former is encountered also in certain fatty acids and derived esters, while the basic four-atom ring of azetidine occurs in some antibiotics, including nocardin and the penicillins. The terminal CH_2 group of the cyclopropyl amino acids and of γ-methyleneglutamic acid has no parallel in protein amino-acid structures.


Azetidine-2-carboxylic acid


β-(Methylenecyclopropyl)alanine
(Hypoglycin A)

OCCURRENCE AND DISTRIBUTION

General statements regarding the occurrence and distribution of the acids are subject to two important limitations. Firstly, the plants investigated represent only a small proportion of the plant kingdom; indeed, very few families have been surveyed systematically. Secondly, since the paper-chromatographic procedure has a definite lower limit of sensitivity, quantities of amino acids representing less than about 1 per cent of the quantity of the major free amino acids on a chromatogram may be easily overlooked. Therefore the conclusion that these newer amino acids are not components of protein is probably too dogmatic, and it is, in fact, more accurate to state that whole plant proteins contain not more than about 0·05 per cent by weight of any of the non-protein amino acids. In fact, α-aminoadipic acid has been isolated from the water-soluble protein of maize seeds in which it represents 0·06 per cent of the dry weight. Similarly, sarcosine

$HOOC.(CH_2)_3.CH(NH_2).COOH$
α-Aminoadipic acid

$CH_2(NH.CH_3).COOH$
Sarcosine

(*N*-methylglycine) has been isolated in 0·03–0·04 per cent yield from a peanut-seed protein. If these acids were distributed throughout the whole seed protein, they would provide only one amino-acid residue for every 2000–3000 present in the protein. Therefore it seems more probable that the acids are restricted to one or more individual protein species, for example particular enzymes, in which they would constitute considerably higher proportions of the final molecules.

Although changes in methodology may produce some modification of present views, it is possible to survey the distribution of particular acids among plants. This distribution is very variable. Extreme examples are γ-aminobutyric acid and hypoglycin A. The former seems to be distributed universally—a plant that did not contain it would be considered odd. In contrast, hypoglycin A is known only as a constituent of the species from which it was isolated, *Blighia sapida*. Many other

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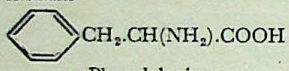
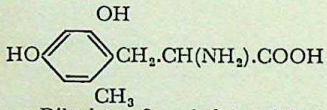
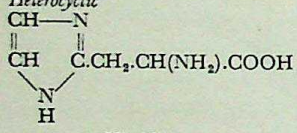
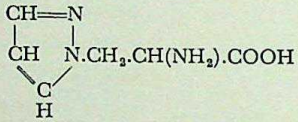
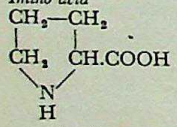
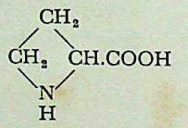
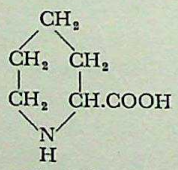
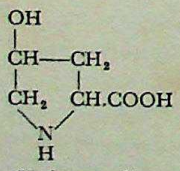
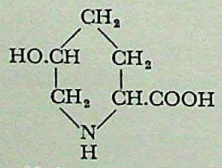
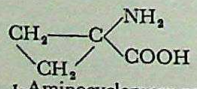
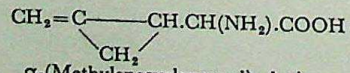
TABLE I

Structural relationships between some protein and non-protein amino acids

Protein amino acid	*Related non-protein amino acid*	*Occurrence in plants*
Acidic		
$HOOC.CH_2.CH_2.CH(NH_2).COOH$ Glutamic acid	$HOOC.C(=CH_2).CH_2.CH(NH_2).COOH$ γ-Methyleneglutamic acid	Peanuts, tulips, and other random occurrences.
	$HOOC.CH_2.CH_2.CH_2.CH(NH_2).COOH$ α-Aminoadipic acid	Many plants.
Amides		
$H_2N.OC.CH_2.CH_2.CH(NH_2).COOH$ Glutamine	$H_2N.OC.C(=CH_2).CH_2.CH(NH_2).COOH$ γ-Methyleneglutamine	Peanuts, tulips, and other random occurrences.
$H_2N.OC.CH_2.CH(NH_2).COOH$ Asparagine	$(C_2H_5)HN.OC.CH_2.CH(NH_2).COOH$ *N*-Ethylasparagine	Cucurbitaceae.
Basic		
$CH_2(NH_2).CH_2.CH_2.CH_2.CH(NH_2).COOH$ Lysine	$CH_2(NH_2).CH_2.CH(NH_2).COOH$ α, γ-Diaminobutyric acid	Solomon's seal (*Polygonatum* sp.).
Hydroxy		
$CH_3.CH(OH).CH(NH_2).COOH$ Threonine	$CH_2(OH).CH_2.CH(NH_2).COOH$ Homoserine	Peas and many other plants.
Aromatic		
 Phenylalanine	 2,4-Dihydroxy-6-methylphenylalanine	Seeds of *Agrostemma githago*
Heterocyclic		
 Histidine	 β-Pyrazol-1-ylalanine	Cucurbitaceae, especially seeds.
Sulphur-containing		
$HS.CH_2.CH(NH_2).COOH$ Cysteine	$CH_3.S.CH_2.CH(NH_2).COOH$ S-Methylcysteine	*Phaseolus vulgaris* seeds; *Neurospora crassa*.
Imino acid		
 Proline	 Azetidine-2-carboxylic acid	Liliaceae.
	 Pipecolic acid	Legumes and many other random occurrences.
 Hydroxyproline	 5-Hydroxypipecolic acid	Dates, ferns, and legumes.
Cyclopropyl acids		
No equivalent	 1-Aminocyclopropane-1-carboxylic acid	Unripe fruits of apple and pear.
	 α-(Methylenecyclopropyl) glycine	Seeds of litchi (*Litchi chinensis*).

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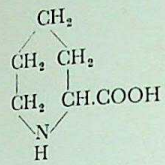
acids have an equally restricted distribution. Another group of compounds, including pipecolic acid, β-alanine, α-aminoadipic acid, and homoserine, have been detected frequently, but not universally, in a range of species that is a fair cross-section of the plant kingdom.

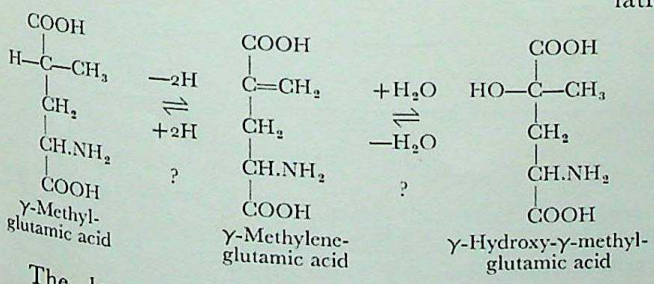


Pipecolic acid

$CH_2(OH).CH_2.CH(NH_2).COOH$
Homoserine

A few acids are characteristic components of particular families. For instance, azetidine-2-carboxylic acid occurs in a high proportion of liliaceous species, but has not been detected in members of other plant families except in a few species of Amaryllidaceae, a family only recently split off from the Liliaceae. Likewise, β-pyrazol-1-ylalanine and *N*-ethylasparagine are known in the cucurbits but not elsewhere. The distribution of γ-methyleneglutamic acid and γ-methyleneglutamine shows a complete contrast, their occurrences being infrequent and haphazard. They are found in unrelated species such as tulips, peanuts, hops, and some ferns.

Chemically related acids frequently occur together in a single species—for example, tulips and peanuts contain combinations of γ-methyleneglutamine, γ-methyleneglutamic acid, γ-methylglutamic acid, and γ-hydroxy-γ-methylglutamic acid, in which the unsaturated acids predominate. There are presumably mechanisms that maintain equilibrium between the last three compounds by the action of enzymes that catalyse hydrogenation-dehydrogenation and hydration-dehydration processes.



The legumes, *Baikiaea plurijura* (Rhodesian teak), and several species of *Acacia*, contain mixtures of pipecolic acid, 5-hydroxy- and 4-hydroxypipecolic acids, and baikiain ($\Delta^{4,5}$-dehydropipecolic acid). Interconversion of these amino acids may depend on similar enzymes. The seeds of the tropical legume, *Saraca indica*, provide a nice example of the coexistence of these two groups of compounds; figure 2b shows that they contain appreciable quantities of γ-methyleneglutamic acid, γ-methyleneglutamine, and pipecolic acid, and possibly smaller amounts of 5-hydroxypipecolic acid.

There are sometimes large differences in the concentrations of particular acids in different organs of the same plant. Thus, dormant seeds of peanut do not contain γ-methyleneglutamic acid, and homoserine is absent from dry pea seeds. Germination is accompanied by a massive production of the respective amino acids, and they become the major components of the free amino-acid pools of the seedlings after a few days' growth. The reverse is encountered in watermelon and cucumber seeds, which contain β-pyrazol-1-ylalanine as the predominant free amino acid. Here, during germination, there is little synthesis of the amino acid, and the leaves of the two plants contain only traces of the acid. These, and numerous other instances of rapidly changing concentrations of non-protein amino acids observed within plants during their development, presumably result from major changes in enzyme activities, and indirectly indicate that such acids may play important roles in intermediary metabolism.

Probably the two most notable accumulations of individual acids are those of γ-hydroxy-γ-methylglutamic acid, which constitutes over 90 per cent of the total non-protein nitrogen of the shoot apices of the maidenhair fern (*Adiantum pedatum*), and azetidine-2-carboxylic acid, which may contain as much as 50 per cent of the total nitrogen present in the rhizome of Solomon's seal (*Polygonatum multiflorum*). Extraordinary accumulations of this type made the original isolations of many of the recognized non-protein amino acids much easier. However, trace amounts of unidentified ninhydrin-reacting substances (presumably amino acids) are detected frequently on chromatograms, and several hundred substances of this type must have been reported in plant extracts. If the normal scale of working were increased tenfold, to use 500 kg quantities of plant materials, it seems certain that a large number of additional amino acids would be isolated and identified.

METABOLISM AND FUNCTION

In most actively growing plant cells, nitrogen metabolism is geared to the production of protein amino acids at the right place and time, and in the

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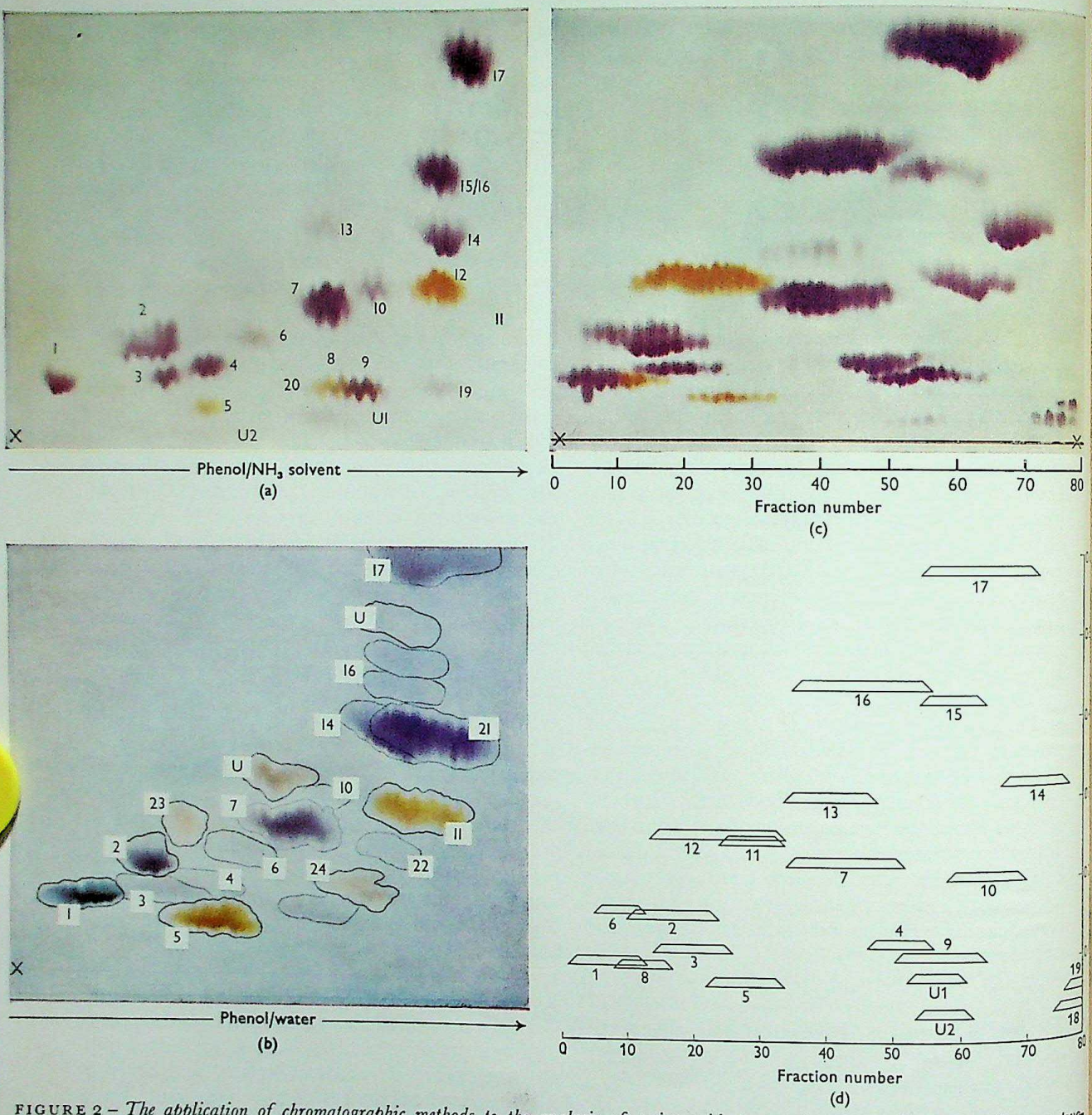

FIGURE 2 – *The application of chromatographic methods to the analysis of amino acids present in plants. Figure 2a shows a two-dimensional chromatogram obtained from an extract of shoots of* Bryonia dioica *(white bryony); Figure 2b is a chromatogram prepared from an extract of* Saraca indica *seed.* X *marks the origin in these two chromatograms. Figures 2c and 2d illustrate the partial fractionation of amino acids obtained when a* Bryonia *extract was treated on a Dowex-50 cation-exchange resin column. Amino acids were eluted by 0·2 N NH_3 solution. In figure 2c* x—x *is the original line of spots. Key to amino acids:* 1, *aspartic acid;* 2, *glutamic acid;* 3, *serine;* 4, *glycine;* 5, *asparagine;* 6, *threonine;* 7, *alanine;* 8, N-(2-*hydroxyethyl*)*asparagine;* 9, *citrulline;* 10, β-*alanine;* 11, *proline;* 12, N-*ethylasparagine;* 13, *tyrosine;* 14, γ-*aminobutyric acid;* 15, *methionine;* 16, *valine;* 17, *leucine and isoleucine;* 18, *lysine;* 19, *arginine;* 20, *glutamine;* 21, *pipecolic acid;* 22, *probably* 5-*hydroxypipecolic acid;* 23, γ-*methyleneglutamic acid;* 24, γ-*methyleneglutamine. U1, U2, and spots labelled U are unidentified. Professor F. C. Steward kindly supplied figure 2b, which also appears in the Ph.D. thesis (Cornell University) of Hugo L. Barrales.*

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correct relative amounts, for their introduction into biologically specific protein molecules. The indispensable character of protein in all phases of cellular metabolism confers automatically a *raison d'être* upon the protein amino acids. No such common function is known for the non-protein amino acids and it is possible that members of this heterogeneous group of compounds play numerous different roles.

Intermediary metabolites

Some of the compounds are implicated in the pathways of biosynthesis and biodegradation of certain protein amino acids [10]. For instance, homoserine is an obligatory intermediate for the production of both threonine and methionine from aspartic acid. γ-Aminobutyric acid, produced by decarboxylation of glutamic acid, may be metabolized further to give succinic semi-aldehyde and succinate; this sequence of reactions provides an alternative to the normal scheme for conversion of glutamic acid into succinate via α-ketoglutarate. β-Alanine may be produced by an analogous decarboxylation of aspartic acid. Independent studies, using *Escherichia coli*, *Torulopsis utilis*, *Neurospora crassa*, and several higher plants, have established that α-aminoadipic acid, pipecolic acid, and α, ε-diaminopimelic acid are all involved in lysine metabolism.

Storage and transport

During periods of less active growth, the orientation of nitrogen metabolism towards protein synthesis is less marked, and in old leaves and in storage organs, particularly during winter months, an overall breakdown of protein may occur. In most plants, much of the protein nitrogen that is released ultimately finds its way into the amides asparagine and glutamine, which are often termed nitrogen-storage compounds. It seems reasonable to ascribe a similar role to azetidine-2-carboxylic acid in many liliaceous plants, where the concentration of this imino acid greatly exceeds that of asparagine and of glutamine. Certainly, the seasonal variations in the azetidine-2-carboxylic acid concentrations in the root and the fleshy storage rhizome of *Convallaria majalis* (lily-of-the-valley) are what one would expect for a compound serving as a nitrogen reserve. *N*-Ethylasparagine, a major constituent of several cucurbitaceous plants, may be viewed as an alternative storage form of asparagine.

Seeds form another type of storage organ, but the level of free amino acids in these is usually low. However, substantial accumulations of some of the non-protein amino acids occur in seeds; for example, pipecolic acid in beans. In the early stages of seedling growth, nitrogen present in these acids probably becomes available for the synthesis of more vital nitrogenous constituents.

$HOOC.CH(NH_2).CH_2.CH_2.CO.NH_2$
Glutamine

$HOOC.CH(NH_2).CH_2CO.NH_2$
Asparagine

$HOOC.CH(NH_2).CH_2C(=CH_2).CO.NH_2$
γ-Methyleneglutamine

$HOOC.CH(NH_2).CH_2.CO.NH(C_2H_5)$
N-Ethylasparagine

Nitrogen is translocated within plants in the form of small soluble molecules, of which asparagine and glutamine are amongst the most important. In peanut plants, γ-methyleneglutamine appears to fulfil this role—over 95 per cent of the total nitrogen present in sap exuding from cut stems is in the form of this amide.

Biogenesis

At present there is insufficient information to give a comprehensive account of the biosyntheses of all the new amino acids, and it is not clear whether they are metabolic by-products or not. The synthesis of any one acid may be catalysed by specific enzymes manufactured for the particular purpose, or may be non-specific and due to the action of enzymes whose normal role is to produce a structurally-related protein amino acid.

Transamination reactions, in which appropriate keto acids accept amino groups from such primary products of ammonia assimilation as glutamic acid, may represent the final step in the formation of many non-protein amino acids. These transaminase enzymes exhibit high degrees of selectivity, but frequently do not exhibit absolute specificity. For example, a purified glutamate-oxaloacetate transaminase from cauliflower buds (this plant does not contain γ-methyleneglutamic acid) will not utilize most amino acids as substrates but will catalyse transamination of γ-methyleneglutamic and γ-methylglutamic acids. The latter acids, however, react approximately a hundred times more slowly with oxaloacetate than does the normal glutamic acid substrate. Since transamination reactions normally are reversible, non-specific enzyme action may be expected to produce these two acids if the appropriate keto acids are available. This type of situation may produce the trace amounts of γ-methyleneglutamic acid detected in pea and bean plants, but the reaction rates are

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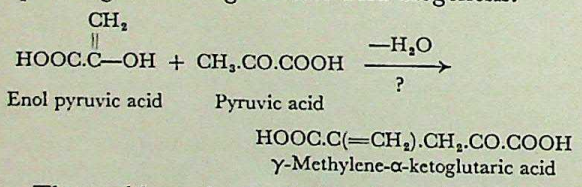
probably too low to account for the large quantities of the acid present in peanut plants. This is supported by preliminary studies that suggest that peanut plants contain a separate enzyme responsible for transamination of γ-methyleneglutamic acid. The formation of γ-methyleneglutamine from γ-methyleneglutamic acid in these plants also seems to be catalysed by an enzyme distinct from glutamine synthetase, which produces glutamine from glutamic acid.

The keto-acid analogue of γ-methyleneglutamic acid is probably formed by condensation of two molecules of pyruvic acid, a universal constituent of plants that is an intermediate in the respiratory breakdown of carbohydrate. Nothing is known about the enzyme responsible for this conversion. It may be a highly specific enzyme present only in those plants producing γ-methyleneglutamic acid. Alternatively, a widely distributed protein normally displaying some other catalytic activity may provide a surface upon which condensation can occur. If this is so, the cells of most plants must possess some intracellular barrier that prevents pyruvic-acid molecules approaching the protein surface, and thereby makes γ-methyleneglutamic acid synthesis impossible. Condensation of pyruvic acid, and the subsequent synthesis of the amino acid, would happen only where a 'metabolic accident' had led to the breakdown of this barrier. Thus we see that the synthesis of γ-methyleneglutamic acid may be controlled by a factor operating at the stage of keto-acid biogenesis.



The problem of γ-methyleneglutamic acid synthesis has been treated in some detail because it illustrates many of the difficulties encountered in attempts to elucidate the biosynthetic pathways for these little-known amino acids. A similar situation is found in the synthesis of pipecolic acid; in *Neurospora crassa*, and in pea and bean seedlings, enzymes involved in proline synthesis catalyse the production of pipecolic acid. However, these non-specific reactions may not proceed rapidly enough to account for pipecolic acid production in all plants.

Control of metabolism

Closely related compounds often exhibit metabolic interactions and many of the most effective enzyme inhibitors are substances bearing a structural relationship to the normal substrate. Certain of the non-protein amino acids may act as regulatory substances in the synthesis and degradation of protein amino acids and hence indirectly affect protein turnover. Examples of such action have been found in the inhibition by γ-methyleneglutamic acid of glutamic acid oxidation in mitochondria from peanut seedlings, and in the reduced rate of incorporation of proline into the protein of carrot-root tissue by azetidine-2-carboxylic acid. It would be interesting to know whether such interactions have any wider significance in the control of differentiation and growth.

CONCLUSION

The last fifteen years have been sufficient to uncover a whole new group of natural products, but this time has been far too short to solve the multitude of metabolic and functional problems raised. One of many such problems concerns the mechanisms by which plants form intriguing structures like the pyrazole, cyclopropane, and azetidine ring systems. The versatility of plants in effecting complex biosyntheses certainly outstrips man's ingenuity in suggesting rational ways in which these structures are elaborated or in providing reasons why they should exist at all.

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Early meteorological observations and the study of climatic fluctuation

GORDON MANLEY

There appears to be a pattern to the fluctuations of the climate of, for example, Europe, but in order to discern this pattern, it is necessary to be able to relate early meteorological observations to each other and to those of the present day. This article describes a number of the early recordings and discusses the errors of the instruments with which they were made. It also discusses the patterns of climatic variation that seem to be revealed by the records.

European civilization developed in the Mediterranean lands. From there it spread northward by the coastal sea routes and the inland river routes, always towards greater climatic obstacles. To the north-west, cooler summers meant decreased evaporation, and too much rain became a problem. To the north, the variable character of spring, with its irregular incidence of cold dry wind and damaging frost, was serious. To the north-east lay an increasing severity of winter. Beyond the isotherm of 0° C for January, snow cover was persistent, and rivers commonly froze. In cultivable lowlands, the Roman Empire was approximately limited to areas with an annual average of no more than 30 days of snow-cover.

An immense amount of the unique technical achievement of Europe stems ultimately from the need to provide for climatic vicissitudes. The ambition of European man has for ever been to free himself from the grinding toil and anxiety of peasant agriculture. The harnessing of the energy of the wind, of running water, and of fossil fuels, to drive machines capable of supplementing or replacing muscle, human or animal, makes a familiar story. Why all this took place in Europe more rapidly than in any other part of the world, for example China, remains a perennial subject of discussion.

Ever since the waning of the last Ice Age, the northward drift of European man has inevitably given rise to his long-standing and quite fundamental concern with the weather. Long before the beginnings of instrumental measurement of the properties of our atmosphere, we have evidence of the dawn of scientific accomplishment in the systematic maintenance of daily records of observed phenomena and of the wind direction. Much of our understanding of the weather derives from such observations kept over considerable periods of time in every country of Europe. This requires a peculiar combination of keenness, self-discipline, precision, and capacity for organization. Such attitudes were not wanting in other temperate lands such as China and Japan, but it was, in fact, in Europe that instrumental measurement started, in the seventeenth century.

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Was born in 1902 and was educated at Manchester University and Caius College, Cambridge. He became an assistant lecturer at Birmingham University in 1926, and in 1928 head of the department of geography at Durham University. He was a University Demonstrator and Lecturer at Cambridge from 1939 to 1948, when he became Professor of Geography at Bedford College, University of London. He has published extensively on British and Polar climatology and on the history of cartography.

FLUCTUATIONS OF CLIMATE

That climatic fluctuations of appreciable magnitude occur is now becoming very evident. Their amplitude is small, and we are not confident of their length; but a predominance of cool unsettled summers, for example, can undoubtedly show its effect on the welfare of a generation. During the present century, the decided amelioration of climate around the northern Atlantic, with the associated retreat of the mountain glaciers, has become conspicuous. The economic effects on fisheries, on forest growth, and on the navigability of Arctic waters have been notable: and it is a question of importance, especially in Scandinavia, whether, and when, a deterioration will follow. There is growing recognition that significant variations in the amount and seasonal incidence of rainfall occur elsewhere in the world, and that these may perhaps be linked, through the general circulation of the atmosphere, with the recognizable fluctuations, perhaps up to half a century

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or more in length, that affect north-west Europe. There is, for example, some evidence of an amelioration early in the sixteenth century, followed by a considerable recession that began to set in some time after 1550. The latter may perhaps be linked with a spell of dry years in the American south-west, but we cannot yet be sure.

The scrutiny of every form of record of climatic vicissitudes in historical times has understandably become very widespread. In the search for precision, we can especially make use of the contributions made by the founders of meteorological science and by their predecessors, who simply recorded events. It is in the lands around the North Sea that we can most clearly trace the progress of meteorological observation and make use of the results.

Something indeed can be made of the many scattered notes of outstanding meteorological events that were recorded by the chroniclers during the centuries before scientific instruments were invented, but it is not easy to establish reliable data. Statements that, in a given winter, the Seine or the Thames was frozen, ignore the possibility of changes in the breadth and depth, or in the obstacles that affected the flow in the past. Statements about the ripening of crops involve consideration of differences between medieval farming practice and that of today.

Here and there in Europe, towards the end of the sixteenth century, we begin to find journals of some length, in which daily notes of wind direction and weather are sufficiently consistent to enable us to attempt estimates of the extent to which conditions approximated to those of today. The stage was set for the invention of meteorological instruments; the seventeenth century saw the invention of the thermometer, barometer, anemometer, and hygrometer, and the founding of the first scientific societies. The earliest daily instrumental observations that we know of were kept in Florence in 1655. Observations were made in Paris in 1658; and Sir Robert Southwell is said to have been the first to introduce to England, in 1659, the newer type of Florentine thermometer.

In the age of Huyghens, Newton, and Leibnitz, the recording of fluctuations with time was an obvious development, and it is appropriate for Englishmen to note how very soon it was after the foundation of the Royal Society that Dr Wilkins read a paper, in which he advocated the keeping of a 'history of the weather'. As a result, Robert Hooke, the Curator of Experiments, was asked to devise a plan for its daily observation, and on 7th October 1663 an account of this was read.

ROBERT HOOKE AND THE FOUNDATIONS OF SCIENTIFIC METEOROLOGY

Hooke's contribution to meteorology was outstanding, and, thanks to some recent discoveries of hitherto unknown meteorological journals, it is now possible to provide for the London region a continuous series of daily observations from 1668 onwards. These link Hooke's pioneer work with the records kept by the meteorologists of the present day.

Hooke presented 'A method for making a history of the weather' with an illustration of his 'scheme of the observables' (figure 2). Hooke's 'method' is said to have been sent to a number of keen observers, including men such as John Beal of Yeovil, in Somerset, who is known to have kept observations, that have since been lost, for many years. Of the other early Fellows of the Royal Society, perhaps the most notable of those who were influenced by Hooke's scheme was John Locke. Locke's first meteorological journal, kept at Oxford from June 1666 to March 1667, is the earliest instrumental record of any duration that we have.

Hooke's 'scheme' set a standard that was followed, at least in part, by his successors for sixty years or more. Indeed, his recommendation of a folio notebook, ruled in nine columns containing the day and hour of observation, followed by 'the strength and quarter of the wind; the degree of heat and cold in the air, best observed by a sealed thermometer; the degrees of dryness and moisture in the air; the degrees of pressure; the constitution or face of the Sky and Heavens . . . whether these clouds afford fogs and mists, or sleet, or rain, or snow . . . whether the way they drive is the same with the wind that blows below . . .' is such that even today the routine of entries made by many observers could be entered under Hooke's headings without difficulty. We now have added measurements of precipitation, and a much more precise notation for the recording of eye observations of the state of the sky, the visibility, and the weather; but the essential principles were thought out nearly 300 years ago. Hooke's recognition of the need for some definition of terms regarding the state of the sky was also far ahead of his time.

Having devised his method, Hooke was requested, on 4th January 1665, to produce a thermometer that should serve as a standard;

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he did this at a meeting on 11th January. He used 'tincted spirit of wine'. The glass tube was graduated above and below a zero at the freezing-point of water, on a scale using 'degrees' that a few years later he halved. The value of his later degree has been worked out from other evidence by Louise D. Patterson [1] as approximately 2·4° C, or 4·3° F; she shows how widely this 'Royal Society's thermometer' scale became recognized as a standard. Hooke, in his 'Micrographia', describes his method of graduation, according to which his scale divisions corresponded to a given increase in volume. Average weather in summer sent the reading up to 8 of his degrees above zero. This can be seen from the entries in his diary, now at the Guildhall Museum, London, in which meteorological observations run from March 1672 to May 1673; there are also occasional later entries. An edited version of this diary was published in 1935.

During the next fifty years, thermometers using this scale were made by a variety of craftsmen, including Tompion himself. Some, however, placed the zero of the scale at an intermediate mark called 'temperate'—about 52° F. Such an instrument is described in a trade card of about 1685 by John Warner, to be seen in Trinity College Library at Cambridge. Commonly the thermometers were quite large, between two and three feet long, with a spherical bulb two inches or so in diameter. As the spirit was not standardized and the glass not annealed, the instruments were unreliable; quite a variety of errors are found when the observations are carefully examined.

THE NEED FOR CAREFUL INTERPRETATION

Of all the meteorological elements commonly measured, the temperature of the air is probably that of the greatest concern to man. Any variations that have occurred in the past require elucidation, because of the demonstrable significance of quite small changes. For example, in southern England or Holland, a rise in the summer mean temperature of 2° C would probably make it possible to start the commercial production of wine; a fall of a like amount would make it uncertain whether wheat-growing on an extensive scale would yield a profit.

Everywhere in the populated lowlands of western Europe, summer evaporation slightly exceeds the rainfall. But with a slight rise of temperature, evaporation increases and rainfall commonly decreases. There is evidence that, in the past 5000 years since the post-glacial 'Climatic Optimum', the range of variation of the mean summer temperature of any given century has probably lain little, if at all, outside the above limit, that is within about 2·5° C on either side.

Hence elucidation of the reasons for the variation shown within the past 300 years may well throw light on the greater variations of the past. Further, we can then link the studies of temperature variation with those of the distribution of pressure, and of the mean direction and strength of the wind. This last becomes important because early barometer readings are also suspect. But it will be evident that if small fluctuations, of about 2° C in the mean temperature over three or four decades, can be so very significant, great care must be taken to compare the results given by these primitive instruments with those we obtain today.

Interpretation indeed is formidably difficult, for before 1750, instruments were few and precious, and in England both thermometer and barometer were generally kept indoors, beside a window, or in some position free from direct radiation, either from the sun or from a fire. Their readings do not differ so much as one might expect from those made in the open air. Contemporary chimneys were large, and evidently English houses were often very cold and draughty. Locke, in a sitting-room of the well-built country house in Essex to which in his old age he retired for most of the winter and spring months, frequently recorded morning temperatures below 40° F. More than a hundred years later, Gilbert White is one of several men who remind us in their journals of that favourite eighteenth-century criterion of severity—ice formed overnight within the bedroom.

Hooke's ingenuity bore fruit in meteorological instruments other than the thermometer. In 1664 he devised a simple type of pressure-plate anemometer, and in 1669 his 'wheel barometer', whereon the movements of the mercury were transmitted to a pointer on a dial (figure 3). He does not appear to have shown so much interest in rain-gauges. It was in the wetter parts of Lancashire, at Burnley, that Towneley began measuring rainfall in 1677, and the ingenuity with which he led the rain down from a funnel on the roof to a position convenient for reading is noteworthy. He could not then have known that such a location for a gauge gives unsatisfactory results. Nearly a hundred years elapsed before Heberden's experiments, using the tower of

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Westminster Abbey, showed that the catch at an elevation was likely to be less than that measured at ground level.

Early barometer readings suffered from the imperfection of the Torricellian vacuum and accordingly tended to be too low, and are not easily comparable; more than a century elapsed before it became the practice to boil the mercury in order to drive out the air. Seventeenth-century scientists soon perceived these imperfections, but, as they had no agreed standard for comparison of observations, progress in technique was necessarily slow.

SOME COMMENTS ON THE EARLY CONTRIBUTORS

The principal early contributors whose meteorological journals can be made use of include John Locke, the philosopher, who left us several lengthy runs of observations kept at various places and times between 1666 and 1703. Robert Hooke's own London journal, is another source. Following these we have 'instrumental' journals in manuscript by J. A. Conyers (1673–81), J. Downes (1680–94), H. Hunt (1692–93), and W. Derham (1697–1706), together with non-instrumental journals, notably those of J. Gadbury (1668-1700), an anonymous manuscript journal at the Bodleian Library (1699–1717), and of J. Smith (1713 onwards to 1745). These enable us to link the work of Hooke with the series of daily instrumental observations at London that is almost complete from 1723.

Some comment on these seventeenth- and eighteenth-century contributors, apart from Hooke and Locke who are well known, may well be made; interesting light is shed on the prevalence of the type of mind that became inspired to purchase instruments and to set about the self-imposed task of daily observations over lengthy periods of time. Most of the observers figure in the Dictionary of National Biography; John Downes and William Derham were the most noteworthy. Downes was physician to Christ's Hospital; he had graduated at Leyden in 1657, and we may surmise that his interest in recording the weather began there. It is noteworthy that in the eighteenth century Dr James Jurin, who, after becoming Secretary of the Royal Society, reiterated in 1723 [2] the desirability of maintaining daily meteorological observations, had been a pupil at Christ's Hospital (1694 onwards) and was also a graduate of the Leyden medical school, at which Boerhaave was emphasizing the importance of observing the weather in order to determine its relationship with the incidence of disease. Jurin's recommendations were followed by a number of English doctors, several of whom were Leyden-trained, for example Huxham of Plymouth and Short of Sheffield. Jurin's own daily observations from 1728 to 1750 provide one of the most important links in the London sequence.

John Downes (1638–1710) became a Fellow of the Royal Society in 1667, but is recorded as 'expelled' in 1675, perhaps for not paying his subscription. His temperature observations begin in 1680 and are noteworthy as they reveal the imperfections of his instrument. Quite evidently the readings changed with time, by something of the order of 8° C in ten years. This seems very remarkable; but we have no equally long contemporary series of observations with which to make comparison. It can be judged, from scattered readings by other observers, that the change did not take place uniformly over the period. The zero of the instrument corresponded to about 2° C in 1680, rising to 8° C by 1683 and 10° C by 1691. There is no indication that Downes ever compared his instrument with others. Shrinkage of the bulb appears to have been a likely reason for the change. Downes was evidently well in touch with contemporary science, and his technique of observations was so much like that recommended by Jurin thirty years later that we may surmise that Jurin as a schoolboy had acquired his ideas from Downes.

Conyers was an apothecary who appears also to have been on the fringe of contemporary science, but his crabbed and difficult manuscript notes give the impression that he was something of a mystic with ideas of his own about humidity.

Other information can be gained from students of astrology, which, moreover, was still a more or less respectable, and gainful, pursuit. Two of its prominent seventeenth-century practitioners contributed material of value to those now endeavouring to reconstruct the events of that period. John Goad, in his 'Astro-meteorologica' (1686), gives many scattered notes on the day's weather which show that he was a keen observer. A more unusual character, perhaps, was John Gadbury (1632–1710), who, under the Commonwealth, was a student of Christ Church. After the Restoration he published almanacs, and in 1691 his 'Astrological Seaman', in which, at a time when marine insurance had begun to develop, he still recommends to merchants, with many examples, the

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importance of casting the ship's horoscope. Whatever may be said of his political views and the numerous oblique remarks in his writings, he was evidently a decidedly accurate observer of daily weather; and in his work of 1691 he provides a journal covering 21 years. In the Bodleian Library, a recently discovered manuscript in Gadbury's hand provides a continuation of this journal to the end of 1700. The consistency of these daily observations is noteworthy, and goes far to assist interpretation of the primitive instrumental observations kept by Downes and Locke.

The last of these seventeenth-century observers, William Derham (1654–1732), was the first of our meteorological pioneers whom we know to have kept his thermometer in the shade and out of doors. The exposure was on the north wall of his rectory at Upminster, fifteen miles east of the City of London. From 1697 onwards, he drew up his daily observations of pressure, wind direction and strength, weather, and amount of rainfall, in a compact table and reported them each year to the Royal Society. At the beginning of 1699, he acquired his thermometer ('nearly three feet long, graduated in inches'), and thereafter he took his readings at fixed hours thrice daily with little interruption. His careful manuscript tabulations for eight years, are available and are remarkably impressive (figure 4). From them the mean change of temperature between his morning, noon, and evening observations can be calculated, and this enables an estimate to be made of the properties, from the standpoint of radiation, of the wall on which his thermometer was hung. Taking this into account, a reasonable estimate of the equivalent air temperature by today's standards becomes practicable. As Derham's observations overlap those of Locke, which in part overlap those of Downes, one can obtain approximate monthly means for London, back to 1680; this includes 'probable' values based on Downes's earlier, very imperfect, thermometer. The trend of these results for the four seasons is shown in the accompanying diagram, which gives the characteristics of the fluctuations of mean temperature in the English Midlands down to the present day (figure 1).

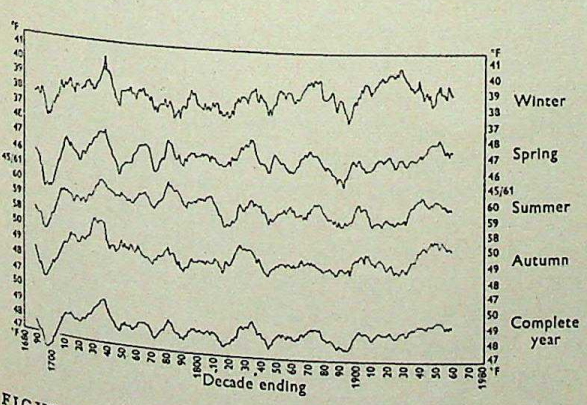

FIGURE 1 – *Averages of seasonal and annual mean temperatures, central England, 1680–1960.*

Derham's observations of the wind, of 'the flying of the clouds', and of the frequency of rain and snow, testify to the fact that no essential change has occurred in the character of our English seasons. This is a matter about which we are sufficiently aware from the frequent, if subjective, comments in the familiar diaries of Evelyn and Pepys.

It might, therefore, be argued that we know enough, and that little advantage accrues from the attempt to relate the material implicit in these primitive and unreliable instrumental readings to modern standards. This argument no longer holds: it will now be shown that these early observations can still serve the purpose of the modern scientist.

CLIMATIC FLUCTUATIONS SINCE 1660

Of late years, interest in climatic fluctuations has developed, largely as a consequence of the work of Scandinavian glaciologists under the leadership of Hans Ahlmann [3]. In Scandinavia, and in the Alps and Rockies, it has been shown that the mountain glaciers as a whole have, during the past few centuries, advanced or retreated at about the same time, and that these changes are largely related to fluctuations in the mean temperature of the summer months. If such a correspondence can be recognized over much of the Northern Hemisphere, the causes may fairly be sought in fluctuations of the general world circulation of the atmosphere, and these in turn may be reflected in events elsewhere, for example, in fluctuation of rainfall on the margin of the subtropical deserts. In the cooler temperate lands of Europe, we can recognize that, in the past, certain decades have been marked by an unusual predominance of unsettled weather accompanied by widespread bad harvests. This was notably true about the period near the end of the Napoleonic wars and about the late 1870s. The 'great droughts', that is, periods of rainfall deficiency, in the western United States may be a manifestation of the same type of fluctuation. Of late, efforts

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have been made, notably by E. B. Kraus [4], to relate the variations in the seasonal incidence of Australian rainfall to events in the Northern Hemisphere.

To elucidate the incidence, the amplitude, and the causes of these fluctuations, which, as far as we know, show no defined regularity in their occurrence, is manifestly desirable. The causes may be solar or terrestrial in origin, or they may be related to the behaviour of the ocean waters and the sea ice; they are potentially of the greatest economic importance. It is in Europe that we possess the longest series of systematic meteorological observations, and it is in England that, starting with the work of Robert Hooke, we may now be able to establish the longest coherent instrumental record of the vicissitudes of the seasons in a critical part of the north temperate zone.

The need for such studies has been widely recognized; for example, C. E. P. Brooks and T. Hunt [5] made a study of the fluctuations in the mean wind direction since 1667. Indeed, efforts to put together lengthy tables of data were made much earlier—by John Dalton, for example, in 1799, and in Germany by Heinrich Dove between 1840 and 1850. James Glaisher, the superintendent of the meteorological observations at Greenwich Observatory, which were initiated in 1840, began to assemble some of the earlier London observations into a coherent series to satisfy the demand created by the current need for information on rainfall, water supply, public health, and the spread of epidemics. Buchan (1893) did the like for Scotland, and also for an independent series of London observations. These have been integrated with Glaisher's earlier data by C. E. P. Brooks, and more recently by B. J. Birkeland [6], to provide a table of monthly mean temperatures representative of the London area from 1763 onwards.

The longest English meteorological record to have been kept in one location and carefully standardized is that for the Radcliffe Observatory at Oxford, started in 1815. Elsewhere in Europe quite a number of national observatories and institutions exist with longer records, and many papers have been published on them; one may mention Oslo, where records start in 1816, Stockholm (1756), Copenhagen (1798), Prague and Vienna (1775), Edinburgh (1764), Paris (1757), Basle (1755), and Leningrad (1743). Still longer tables can only be built up, as a rule, by putting together the results from several neighbouring localities whose records overlap. The most impressive of these was compiled by Labrijn for Utrecht, Holland, beginning in 1706. Berlin has a record with but slight interruption back to 1719; London, as we have seen, can likewise be taken back to 1723 with the aid of Jurin's journal. Across the Atlantic, New York (1822) and New Haven (1780) are noteworthy.

Temperature fluctuations are probably the most important, and there have unfortunately always been changes in the sensitivity of instruments, in hours of observation, and in exposure, as well as in actual location. These have occurred everywhere and must be allowed for. Complex arguments arise because the majority of the earlier European observations were kept in capital cities or in towns that have since grown; this makes it difficult to decide how much of any trend can be described as man-made. Complex arguments also arise with regard to the interpretation of the terms used in description of weather. As England in the eighteenth and early nineteenth centuries was *par excellence* the land of the individualistic amateur, all these problems of reduction become acute. Fortunately, some remarkably long and consistent rural records are available; that kept by Thomas Barker, brother-in-law of Gilbert White, in Rutland from 1733 to 1798 is outstanding. It is through Barker that Glasspoole has been able to discuss the fluctuations of English rainfall back to 1727 [7]. It is interesting to note that in Holland meteorological records can also be carried back to the later seventeenth century, thanks to the existence of the long 'country' series of observations at Zwanenberg from 1735 to 1861.

Fluctuations in the mean distribution of pressure, and in the mean wind direction, point to circulation changes of importance, and have recently been studied by H. H. Lamb of the Meteorological Office, London, who has carried his maps of North Atlantic pressure distribution for each decade back to 1760 [8].

There is considerable evidence that, before that date, north-west Europe was favoured by remarkably dry weather, notably about 1740–43, and by a prevailing amelioration of climate between 1720 and 1740, which has been considered to be very beneficial, for example, to the population of Sweden [9]. There is also abundant evidence that the decade 1692–1701 was unusually unfavourable. It is known that widespread glacier advance set in, culminating in Iceland and in the Alps about 1715. Economic distress in Sweden was

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ROYAL SOCIETY. 179

A SCHEME

At one View representing to the Eye the Observations of the Weather for a Month.

Days of the Month and place of the Sun. Remarkable houſe.	Age and ſign of the Moon at Noon.	The Quarters of the Wind and its ſtrength.	The Degrees of Heat and Cold.	The Degrees of Dryneſs and Moyſture.	The Degrees of Preſſure.	The Faces or viſible appearances of the Sky.	The Notableſt Effects.	General Deductions to be made after the ſide is fitted with Obſervations: As,
14 ♊ 12.46	27 ♂ 9. 46. Perigeu.	W. W.SW.	2. 9 3 12 3½ 16 10 ⅛ 17 ½	3 2 ½ 2 8 2 9 ½ 2	29 1/16 29 ⅛ 29 ½	Clear blew, but yellowiſh in the N. E. Clowded toward the S. Checker'd blew.	A great dew. Thunder, far to the South. A very great Tide.	From the laſt quarter of the Moon to the change the weather was very temperate but cold for the ſeaſon; the Wind pretty conſtant between N. and W.
15 ♊ 13.40	28 ♂ 24. 51.	N.W. N.	3 9 4 2 8 ½ 1 7	2 8½ 2 9 2 10	29 1/16 29	A clear Sky all day, but a little checker'd at 4. P.M. at Sunſet red and hazy.	Not by much ſo big a Tide as yeſterday. Thunder in the North.	A little before the laſt great Wind, and till the Wind roſe at its higheſt, the Quickſilver continued deſcending till it came very low; after which it began to reaſcend,
16 ♊ 14.37	10 N.Moon. at 7. 25' A.M. ♊ 10. 8.	S.	1 10	1 10	28 ½	Overcaſt and very lowring.	No dew upon the ground, but very much upon Marble ſtones,	
	&c.	&c.	&c.	&c.	&c.	&c.	&c.	&c.

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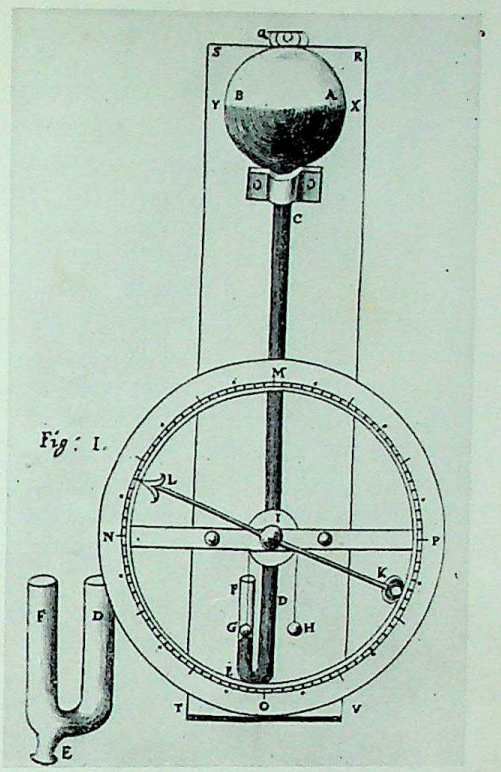

FIGURE 3 – *Robert Hooke's wheel barometer.*

FIGURE 2 (*left*) – *Robert Hooke's scheme of observations.*

	May.			5. 12. 9			June.			5. 12. 9		
	Weather.	Winds.	Clouds	Barom.	Ther.	Rain	Weather.	Winds.	Clouds.	Barom.	Ther.	Rain
1	Fair & pleaſant.	EbN¹		29·68 79 87	87 135 101		Cloudy & Cool.	ENE⁴		29·63 65 71	126 143 124	
2	Same.	EbN⁰		87 96	91 105		Same.	NNE² NE³		69 70 77	118 143 112	
3	Cloudy. Rainy. Cloudy.	NW¹		98 30·06	105 108		Clear Cloudy.	NE¹ NbN¹		77 77 71	98 151 134	
4	Cloudy.	N¹ NNE²		06 06 04	99 114 104		Cloudy. Rain.	SbE¹ SE²	SSW	57 50 47	129 150 126	
5	Same.	N³		29·98 88	103 111		Cloudy.	ENE² NE³		50 57 65	124 145 117	0·33

FIGURE 4 – *Part of William Derham's notebook for May and June 1704. The figures at the top give the times at which the observations were made. (By permission of the Royal Society.)*

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serious, and in Scotland catastrophic. In Iceland, 1695 stands on record as the summer when the sea-ice drifted around the whole island; about 1720, the advance of the Grindelwald glaciers ended. And in regard to frequency of snow in London, 1695 is the only year to rival 1784, 1879, 1917, and 1947. The spring of 1698 in England and the subsequent harvest were probably the most tardy that we know of; and the April of 1701 was quite unusually cold and snowy. The aggregate effect on the trend of temperature can be seen in figure 1.

To provide a quantitative assessment of what happened, we have these early records. Thus we can show, by means of the accompanying table, the marked increase in the frequency of precipitation, both rain and snow, in south-east England and the decided fall in the mean temperature, especially in spring and summer (figure 1). But an appreciable amelioration developed after 1702, following the decade 1692–1701, which appears on the whole to have been cooler than any we have since experienced, although 1809–18 ran it close. While the attempt to reduce such early observations to modern standards is subject to a multitude of chances of error, the overall averages of winter temperature for the years 1680–1706 can be related to the average frequency of snow and compared with the present day; this provides some check on the values reconstructed from these scattered and imperfect instruments. Even if the results might still be regarded as uncertain, the pattern they display assists us to formulate our theories.

Throughout the period 1670–1710, there is little evidence for excessive summer heat, save in June 1676 and on 'Hot Tuesday' (8th July 1707), for which days our instrumental estimates are very sketchy indeed. The extreme cold of the winter of 1684 was outstanding; as a whole it probably exceeded that of any subsequent winter except, perhaps, 1740. This accords well with contemporary reports of the length of time that the Thames remained frozen.

It is possible that the climatic recession of the 1690s was a necessary precursor of the amelioration of 1720–38; we do not know. Neither do we yet know whether it was associated with solar behaviour, save that solar activity, so far as it may be indicated by sunspots, seems to have been very weak. That there were extensive volcanic eruptions in 1693–94 is known, but the series of cool seasons began before that.

The utility of long series of observations lies in the increased opportunity for correlation with events in other parts of the world, such as those which may be revealed in the core-samples taken from the deep oceans [10], in the behaviour of glaciers, or in longer-term fluctuations of rainfall. Moreover, knowledge of the size and duration of earlier climatic recessions and sequences of bad seasons of this type gives us some forewarning of future possibilities.

LONDON WEATHER, 1671–1720

The annual average number of days with precipitation observed and recorded, and of snow seen to fall, is given for each decade. The totals given by different journals have been standardized as far as possible. The present-day average, based on the days with 0·01 in. or more measured, is given for 1921–50.

	Days with precipitation (rain, hail, sleet or snow)	Days with snow observed (and recorded as falling)
1671–1680	155	17·2
1681–1690	167	18·2
1691–1700	179	25·5
1701–1710	168	17·9
1711–1720	165[2]	20·0[2]
	Days (0·01 in. or more)	
1921–1950	168	12·5[1]

[1] By similar standards to those of the early observers, as far as possible.
[2] Approximate.

ACKNOWLEDGMENTS

I have to thank the Officers of the Royal Society for the opportunity to work on the manuscripts left by their early Fellows; and the authorities of the Bodleian Library, to whose manuscript collections we are so greatly indebted.

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The principles of electron-spin resonance

A. CARRINGTON

Paramagnetic molecules have unpaired electrons, and in a changing magnetic field these will absorb energy at particular values of the field. This absorption is due to a change in direction of the magnetic moment resulting from the electron's spin, and the absorption spectra that are produced can give information about the structure of free radicals and complex ions that is unobtainable by other methods. This article describes the theory of electron-spin resonance and some applications of electron-spin resonance spectroscopy in chemistry.

No modern scientist can afford to be unaware of the diverse applications of electron-spin resonance (e.s.r.), first observed experimentally by E. Zavoisky in 1945 [1]. It is restricted to molecules containing unpaired electrons, but this limitation is, in fact, the main reason for its value; the paramagnetic molecules in a complex system can be investigated, and one can gain information that is very difficult to obtain by any other method.

The basic principle of e.s.r. is best understood by considering a single electron. This has an intrinsic angular momentum, called its spin, which has a value of 1/2 quantum unit. The spin quantum number S is therefore 1/2. The spin, M_S, can have two values, +1/2 and −1/2, representing the fact that the spin can be in either of two opposite senses; the spins that have these values are known respectively as an α spin and a β spin.

The electron has a magnetic dipole moment by virtue of its spin. In the absence of any external magnetic field the free electron has no preference for an α or a β spin, as they are of equal energy. When a magnetic field is applied, however, the electron will have lower energy if its spin magnetic moment is aligned so as to be parallel to the applied field, rather than antiparallel, and this will be the preferred alignment. The electron is said, in quantum-mechanical language, to be doubly degenerate in spin when it can exist in two states of equal energy. This degeneracy is removed by a magnetic field (figure 1*a*). When a field is applied, the separation between the two energy levels in which $M_S = +1/2$ and $M_S = -1/2$ is proportional to the product of a constant β (the Bohr magneton) and the strength of the field, H. The proportionality constant, g, is commonly referred to as the 'g-value'. If we irradiate the electron with electromagnetic radiation possessing energy with a quantum value, $h\nu$, equal to the separation between the two energy levels, the electron will absorb energy and 'flip over' to a state of high energy in which its spin magnetic moment is aligned anti-parallel to the magnetic field. The resonance condition is thus represented by the equation

$$h\nu = g\beta H$$

The change in M_S is governed by the selection rule that M_S will be increased or decreased by unity ($\Delta M_S = \pm 1$). Electron-resonance transitions can, clearly, be produced by varying either the magnetic field, or the frequency. In practice the frequency is fixed and the field is changed slowly until it passes through the value required

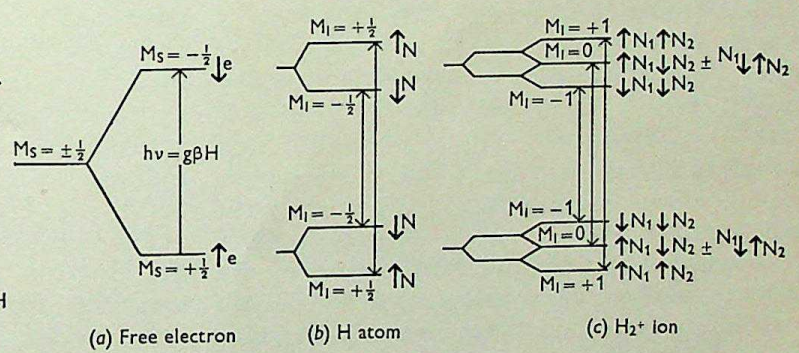

FIGURE 1

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Was born in 1934 and was educated at Colfe's Grammar School and the University of Southampton. He obtained a Research Fellowship in the University of Minnesota, U.S.A., for 1957–58, and was elected into a Research Fellowship at Downing College, Cambridge, in 1960. In 1960 he was appointed Assistant in Research in the Department of Theoretical Chemistry, University of Cambridge. He is at present engaged in research into the electron-spin resonance spectra of aromatic free radicals in solution.

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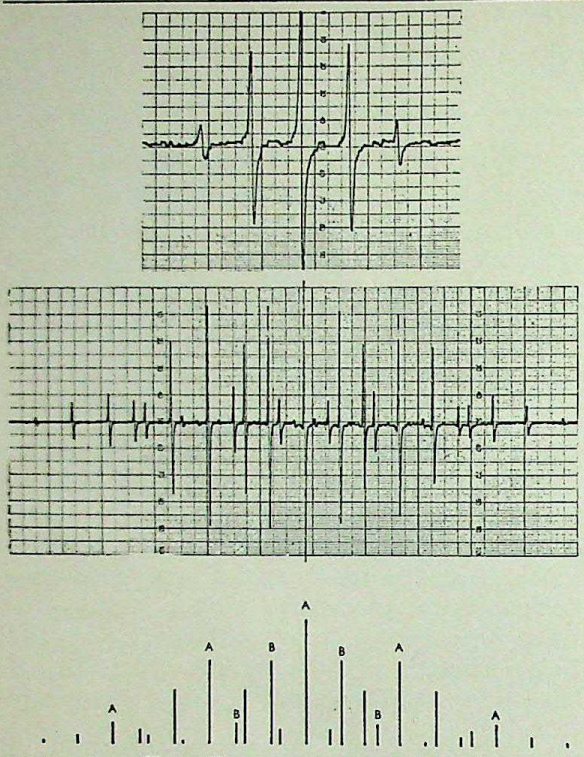

FIGURE 2 – *Top: E.s.r. spectrum of the p-benzosemiquinone ion. Bottom: E.s.r. spectrum of the naphthalene negative ion, with a reconstruction indicating the origin of the hyperfine lines.*

for a resonance transition. Absorption spectra (figure 2) thus represent absorption plotted against magnetic field.

A convenient magnetic field to use in the laboratory is 3000 gauss, and a frequency of about 9000 Mc/s, in the microwave region, is needed to induce electron resonance. The high-frequency radiation is supplied by a klystron and is led through a waveguide to a resonance cavity containing the paramagnetic specimen. If energy absorption occurs, it is detected by a crystal, amplified, and displayed on an oscilloscope or a pen recorder. Resonance cavities can be designed to suit any type of specimen and it is relatively easy to work at different temperatures. Complete e.s.r. spectrometers can now be obtained commercially.

STUDY OF FREE RADICALS IN SOLUTION

For a free electron, g has the value 2·00, and organic free radicals always have g-values very close to this. However, other paramagnetic molecules, such as certain salts of the transition-metal ions, may have g-values different from 2, and, as will be discussed later, measurement of the g-value provides valuable information about the electronic structure of such salts. The g-value often depends, also, on the orientation of the magnetic field with respect to the paramagnetic molecule. A single molecule cannot, of course, be studied, but an assembly of molecules oriented regularly in a single crystal can be. Under these conditions, the measurement of g-value anisotropy is of great interest.

For the moment, attention will be limited to systems in which the g-value is isotropic and has the free-spin value. The hydrogen atom, the simplest free radical, introduces a further possibility. The proton nucleus itself possesses a spin of 1/2, with a resulting magnetic moment, and it prefers to align this magnetic moment parallel, rather than antiparallel, to the electron-spin magnetic moment. Since there are two ways of aligning the electron-spin magnetic moment in an external magnetic field, there are four possible arrangements (figure 1*b*).

As the nuclear spin, M_I, can have a value of $\pm 1/2$, two of the four possible transitions involve no change of M_I, and two of them involve a change of unity. In fact the selection rule is that there shall be no change in M_I, so that only two absorption lines are observed. The resonance spectrum of the hydrogen atom has been observed by several different workers, and the splitting between the lines, the so-called 'nuclear hyperfine splitting', corresponds to a field change of 500 gauss.

For each orientation of the electron spin in the H_2^+ ion, there are three distinct ways of arranging the nuclear spins. These ways are characterized by the M_I values $+1$, 0, -1. The $M_I = 0$ level has two alternative arrangements of spins of equal energy; that is, it is doubly degenerate (figure 1*c*). We therefore expect the e.s.r. spectrum to show three hyperfine lines, of relative intensities 1:2:1 because of these two possibilities for the state in which $M_I = 0$. A system in which the unpaired electron interacts with n equivalent protons will show $n+1$ hyperfine lines, with a binomial distribution of relative intensities. This simple rule forms the basis for the interpretation of the spectra of organic free radicals. One or two examples will illustrate the method. The ionized p-benzosemiquinone radical I, formed by the one-electron reduction of p-benzoquinone in alkaline solution, has four ring protons in equivalent positions. The e.s.r. spectrum of this radical [2] will therefore consist of five equally spaced lines with relative intensities 1:4:6:4:1 (figure 2*a*). A more complicated example is the naphthalene negative ion, II, which

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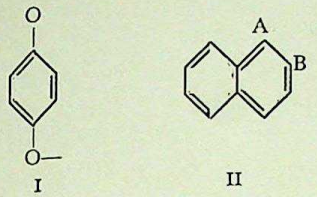
has four protons in the A position, and four in the B (see below).



These two sets of protons give different splittings, for reasons that will be discussed later, and the five lines arising from the A protons are each split into five lines by the B protons. The resulting twenty-five-line spectrum is shown in figure 2*b*; the origin of each line is shown in the guide beneath the figure.

How does this hyperfine splitting arise? The C—C and C—H bonds of the naphthalene molecule are formed from trigonal sp^2 hybrid orbitals on the carbon atoms and hydrogen 1s orbitals. There is thus a 2p orbital, perpendicular to the plane of the molecule, on each of the ten carbon atoms. These ten p_π orbitals can combine with each other to form ten molecular orbitals spread over the whole molecule, five of them bonding and the other five anti-bonding. There are ten p_π electrons, and these just fill the five bonding molecular orbitals. The extra electron in the naphthalene negative ion goes into the next molecular orbital and is thus able to move over the whole molecule. In consequence, there will be a certain 'unpaired-electron density' ρ_1 at each carbon atom, and it has been shown [3] that the hyperfine splitting, a_H, due to a ring proton is directly proportional to ρ_1 at the neighbouring carbon atom. That is, $a_H = Q\rho_1$, where Q is a constant, about 25 gauss. A simple calculation for naphthalene shows that the ratio of the hyperfine splittings due to the type-A and type-B protons should be 2·62:1. The experimental value obtained from figure 2 is 2·68:1. This demonstrates one of the principle uses of e.s.r. spectra. The hyperfine-splitting constants provide a good test of molecular wave functions, and it is encouraging that molecular-orbital theory and valence-bond theory agree with one another.

FREE RADICALS IN SOLIDS

As has been mentioned, the *g*-value is anisotropic in certain cases. This is often true also of the nuclear hyperfine structure, but in the free radicals described, those generated chemically in solution, the anisotropy disappears, in effect, because of the rapid movement of the molecules. The spectrum is, therefore, completely isotropic. However, we can study free radicals oriented in a regular manner, by trapping them in a single crystal, and so can investigate the anisotropy of the e.s.r. spectrum. The method thus has another important application, in the study of damage to organic crystals by high-energy radiation such as X-rays or γ-rays. A typical example is the γ-irradiation of a single crystal of succinic acid, causing the removal of a hydrogen atom to form the radical $(CO_2H)CH_2CH(CO_2H)$ [4]. The e.s.r. spectrum shows hyperfine lines from the —CH and —CH_2 protons, and the separation between these lines varies with the orientation of the magnetic field with respect to the crystal. From a detailed study of this variation, it is possible to determine the hyperfine constants—which can tell one a great deal about the mechanism of the magnetic interaction—and also the orientation of the radical with respect to the undamaged molecules in the crystal.

When the radicals in a solid are randomly orientated, it can be difficult to interpret the e.s.r. spectra correctly, because of the anisotropy of the proton hyperfine structure. For example, γ- or X-irradiated Perspex gives a complicated e.s.r. spectrum that is still not completely understood. In other cases the interpretation is simpler. Thus, ultraviolet irradiation—at the temperature of liquid nitrogen—of a glass consisting of isopropanol, water, and hydrogen peroxide results in the formation of hydroxyl radicals that remove α-protons from the alcohol molecules, leaving the radical $(CH_3)_2C.OH$ [5]. The e.s.r. spectrum consists of seven lines with the expected binomial distribution of intensities, arising from interaction of the unpaired electron with the six equivalent protons of the methyl groups. If the radical could be dissolved, it would be expected to show the same type of spectrum.

Many polymerization processes take place through a free-radical mechanism. Radicals trapped in the polymer are easily detected by e.s.r., and kinetic studies have been made of the increase and decrease of the concentrations of the radicals. For example, the radical concentration during the copolymerization of methylmethacrylate and glycoldimethacrylate has been measured, and its dependence on the constitution of the mixture has been studied [6]. A higher percentage of copolymer is accompanied by a higher radical-concentration, because of increased cross-linking.

Application to biological systems is possible in principle, but not much work has been done. The

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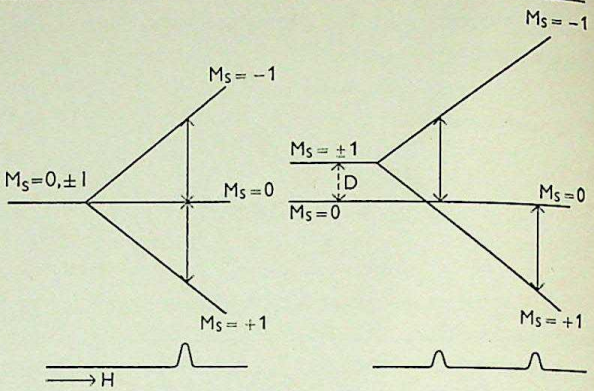
first experiments [7] showed that there are varying free-radical concentrations in living tissues, and, in particular, that the free-radical content is higher in metabolically active tissues, such as green leaf and liver, than in less active ones. An example of uncertain significance, but great topical interest, is the study of free radicals in cigarette smoke [8]. If cigarette smoke is immediately frozen and the condensate examined by e.s.r., strong absorption is observed. On allowing the sample to warm up, the absorption falls to approximately one-sixth of its initial value and then changes little over long periods of time. The inference is that in cigarette smoke there are short-lived active free radicals and long-lived stable free radicals, and it is possible that either or both of these might be carcinogenic. Another important application, referred to later, was to the determination of the structure of haemoglobin.

MOLECULES WITH TWO UNPAIRED ELECTRONS

Let us now consider molecules with two unpaired electrons coupled to form a triplet-spin state. This is so called because there are three ways of arranging the electron spins, each of which spins has the value $\pm 1/2$; M_S will be $+1$, 0, and -1. If the three levels were initially of equal energy, they would be split apart by an external magnetic field (figure 3*a*), and the two allowed electron-resonance transitions, $\Delta M_S = \pm 1$, would occur at precisely the same value of the magnetic field. In fact, the levels are never degenerate, for various reasons, even in the absence of a magnetic field, and consequently the two allowed transitions occur at different magnetic field strengths, and appear as two separate absorption lines. This 'zero-field splitting' is also shown in figure 3*b*. A particularly fascinating experimental example is provided by the phosphorescent state of naphthalene [9]. If naphthalene is irradiated with ultraviolet light, one of the bonding electrons is excited and moves into an unoccupied antibonding orbital. It then changes its spin, and the molecule exists for a time in a triplet-spin state that can be studied by e.s.r. The resonance spectrum of the triplet state shows two lines ($\Delta M_S = \pm 1$) which are each further split by the proton nuclei. This observation has opened up an entirely new field for e.s.r., namely the study of molecules in their excited states. A number of hydrocarbons have been studied, and the results give a picture of the electron distribution in the excited state.



(a) No zero-field splitting (b) Zero-field splitting

FIGURE 3

INORGANIC MOLECULES AND FREE RADICALS

A number of simple inorganic molecules have been studied. These include NO, NO_2, and O_2, studied in the gas phase [10], and ClO_2, studied in solution [11]. The spectra of NO, NO_2, and O_2 are complicated by coupling between the magnetic moment of the unpaired electron and rotational motion of the molecules. It is possible to study this coupling by e.s.r., but the information gained is equally well obtained by other techniques, such as microwave spectroscopy.

So far, short-lived molecular free radicals in the gas phase have not been observed by e.s.r. The technique has been more successful for atoms, and hydrogen, nitrogen, oxygen, and phosphorus atoms produced by high-tension electrical discharges have been studied in the gas phase [12]. Iodine atoms, produced by ultraviolet irradiation of iodine vapour, have also been detected in the gas phase [13].

Irradiation damage to inorganic crystals has been extensively studied by e.s.r. F-centres, which result when a negative-ion vacancy is occupied by an electron, have been studied in irradiated KCl, LiF, NaF, and many other simple inorganic salts [14]. The electron interacts with the surrounding nuclei to produce a complex hyperfine structure that can be resolved and interpreted in detail. V-centres, resulting from removal of a cation from the lattice together with an electron from a neighbouring anion, have also been detected [15]. One of the earliest measurements was on the Cl_2^- ion in X-irradiated KCl; the spectrum shows a small *g*-value anisotropy, and strong anisotropy of the hyperfine structure arising from the two chlorine nuclei.

TRANSITION-METAL IONS

The method is of great value in the study of

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transition metals, which are characterized by having unpaired electrons in d-orbitals. So far, systems have been discussed for which the g-value is isotropic—that is, independent of the orientation of the magnetic field with respect to the paramagnetic molecule—and close to the free-spin value. The transition-metal ions do not always have isotropic g-values, as the unpaired electrons often have orbital angular momenta in addition to spin angular momenta. In general, so far as the transition-metal ions are concerned, an electron will possess orbital angular momentum if there are a number of d-orbitals of equal energy available to it. This situation is commonly met in symmetrical complexes; measurement of the g-value often enables one to identify the d-orbitals concerned. For example, the Ti^{3+} ions possess one 3d electron, and in a complex like $CsTi(SO_4)_2.6H_2O$, where the Ti^{3+} ion is surrounded by an octahedron of water molecules, there are three d-orbitals of equal energy available to the unpaired electron. It will thus have some orbital angular momentum, and the g-values observed are considerably less than 2·0 [16]. The subject is complex and the reader desiring more details should consult the specialized literature [17, 18].

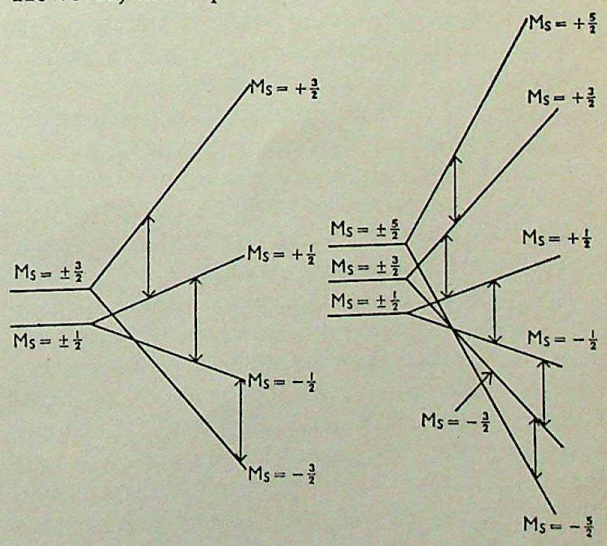
An ion in which the unpaired electron has a number of d-orbitals of equal energy available is said to have an orbitally degenerate ground state. As we have seen, measurement of the g-value distinguishes between degenerate and non-degenerate ground states. Ions possessing several unpaired electrons always show a zero-field splitting between the various spin levels, and a general rule about this can be stated. If the ion has an orbitally degenerate ground state, the zero-field splitting is very large. For ions possessing an even number of unpaired electrons, this may put electron-resonance transition outside the available frequency range. For example, the V^{3+} ion has two unpaired electrons, and, in an octahedral complex, has a threefold orbital degeneracy. The spin triplet is split by spin-orbit coupling in the manner indicated in figure 3, but the splitting is at about 8 cm^{-1}. As the wave number of microwave radiation is usually from 0·3 cm^{-1} to 1·5 cm^{-1}, the levels are much too far apart for it to be possible to induce transitions between them.

On the other hand, a tetrahedral ion with two unpaired electrons has an orbitally non-degenerate ground state. The zero-field splitting is small in such cases (approximately 0·1 cm^{-1} in the tetrahedral FeO_4^{2-} ion with two unpaired electrons [19]) and the two e.s.r. transitions ($\Delta M_S = \pm 1$) illustrated in figure 3b are observed.

Another example of an orbitally non-degenerate ground state is the octahedral Cr^{3+} ion. This has three unpaired electrons, which add up to a total spin of 3/2, and in consequence the spin, M_S, can take the values +3/2, +1/2, −1/2, −3/2. There are three possible allowed electron-resonance transitions for which $\Delta M_S = \pm 1$. If the spin system had the full four-fold degeneracy in the absence of a magnetic field, the three transitions, having the same energy, would occur at the same value of the applied magnetic field. In fact, there is always a small zero-field splitting, and figure 4 shows how three electron resonance lines will be observed for a zero-field splitting between the levels ±1/2 and ±3/2. Similar considerations lead to the general conclusion that if an ion has n unpaired electrons ($S = n/2$), n electron-resonance transitions are possible, although not necessarily observable.

One further example will help to demonstrate this. The Mn^{2+} ion in octahedral or tetrahedral complexes usually has five unpaired electrons. There are five d-orbitals available, so the electrons form a non-degenerate ground state. For such a system, $S = 5/2$, and the spin takes values from +5/2 and −5/2. The ion usually has a small zero-field splitting of the type shown in figure 4b, and five electron-resonance transitions are observed.

The multiplicity of spectral lines that arises from different transitions in which $\Delta M_S = \pm 1$, is described as the 'fine structure' of the spectrum. However, the unpaired electron can interact with



(a) S = 3/2 with zero-field splitting (b) S = 5/2 with zero-field splitting

FIGURE 4

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the nucleus in transition-metal ions, and hyperfine structure is then observable. If the metal nucleus possesses a spin quantum number I, a given electron-resonance line will be split into $2I+1$ lines of equal intensity and spacing, as there are $2I+1$ quantized orientations of the nucleus with respect to the electron-spin magnetic moment. For example, the Mn^{55} nucleus has a spin $I=5/2$; hence the five-electron resonance lines described above are each split into six hyperfine lines. As with free radical spectra, the full selection rules are $\Delta M_S = \pm 1$ and $\Delta M_I = 0$. Again, the magnitude of the hyperfine splitting, that is, the separation between the hyperfine lines, depends on the amount of time the unpaired electron spends near the nucleus. Thus, if the unpaired electrons move in molecular orbitals compounded of metal d-orbitals and ligand p-orbitals, they spend less time in the vicinity of the metal nucleus and consequently the hyperfine splitting is reduced [20]. This illustrates how e.s.r. measurements can be informative about covalent bonding in transition-metal complexes.

Covalent, or 'charge-transfer', bonding is, however, demonstrated even more directly if both the metal and ligand nuclei have non-zero spins, for then both the metal and the ligand atoms will give hyperfine structure. The first ion for which this effect was demonstrated was $IrCl_6^{2-}$ [21]. The Ir^{4+} ion has one unpaired electron, and there is one electron-resonance transition, which is split into four hyperfine lines by interaction with the iridium nucleus ($I=3/2$). However, each of these four lines is further split by the chlorine nuclei, and from the relative magnitudes of the splittings it is possible to determine how much time the electron spends near the iridium nucleus and how much near the chlorine nuclei. Many other salts of transition-metal ions show ligand hyperfine structure, including even the simple divalent fluorides of ions like Mn^{2+} and Co^{2+}.

We have already pointed out that if one wants to measure the anisotropy of an e.s.r. spectrum it is necessary to work with a single crystal. This is not the only requirement, however. In addition, it is usually necessary to study a 'dilute' single crystal,—that is, a mixed crystal of the paramagnetic salt with an isomorphous diamagnetic salt—in order to isolate the paramagnetic ion from similar ions with which it would interact. Several types of magnetic interaction between paramagnetic ions can be distinguished, and a particularly interesting example of 'exchange interaction' is observed in cupric acetate crystals [22]. The cupric ion has one unpaired electron, and one normally observes an e.s.r. spectrum consisting of one electronic transition ($\Delta M_S = \pm 1$) split into four components by the ^{63}Cu and ^{65}Cu nuclei (both with $I=3/2$ and almost equal moments). In the case of cupric acetate, however, the spectrum shows a triplet-spin state with a small zero-field splitting. It was inferred that the copper atoms are situated in close pairs in the crystal, the paramagnetic unit containing two Cu^{2+} ions with their unpaired electrons coupled. As the temperature is lowered, these electrons pair, and this is indicated by a decrease in intensity of the spectrum. This deduction about the crystal structure of cupric acetate has been fully confirmed by X-ray crystallography.

This is by no means the only example of an e.s.r. spectrum being used to determine certain features of a crystal structure. A particularly elegant application of this kind was to the haemoglobin and myoglobin molecules and their derivatives [23]. The central parts of these molecules consist of a porphyrin plane containing an iron atom at the centre of the four nitrogen atoms. A line through the iron atom perpendicular to the porphyrin plane is an axis of symmetry, and when the external magnetic field is along this axis a minimum g-value of 2·0 is observed; in any direction perpendicular to this axis, however, the measured g-value is 6·0. Knowing this, one can proceed to study single crystals of haemoglobin or myoglobin and determine the orientation of the porphyrin planes with respect to each other and the crystallographic axes.

CONCLUSION

We have described a large number of applications of the method, but the list is by no means comprehensive. In conclusion, it is appropriate to consider the limitations of the technique and its relation to other spectroscopic and magnetic methods. Firstly, e.s.r. is never likely to replace magnetic susceptibility measurements; the two techniques complement each other, and it is indeed always desirable to have susceptibility data before commencing a detailed e.s.r. sudy. The resonance results for haemoglobin, for instance, could be very misleading were it not known in advance, from susceptibility measurements, that the iron atom has five unpaired electrons. On the other hand, if one has a transition-metal ion in a diluted single crystal, e.s.r. measurements provide a wealth of detailed information about the energy levels that susceptibility measurements cannot

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give. Not every inorganic compound can be produced in the form of large diluted crystals, and there are other phenomena that may make resonance studies difficult or even impossible, such as a very large zero-field splitting. E.s.r. and visible or ultraviolet absorption spectroscopy complement each other, the latter providing information about excited states that is, at best, obtained only very indirectly from resonance studies.

It is in the free radical field that the technique is virtually unique, though here, too, there are limitations. With the best available commercial equipment, radicals in solution can be detected at a concentration of about 10^{-9} M. In practice, to obtain a reasonably well resolved spectrum, a considerably higher concentration (10^{-6}–10^{-7} M), is used, and in aqueous solutions the sensitivity is rather lower. If identification of free radicals is the aim, a well-resolved spectrum is essential and this is not always easy to obtain.

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Book reviews

SCIENTIFIC EDUCATION

The Complete Scientist. Report of the Leverhulme Study Group to the British Association for the Advancement of Science. *Pp. xxiii+162. Oxford University Press, London.* 1961. 18*s. net.*

This survey of the education of scientists and technologists in Great Britain by a Leverhulme study group under Sir Patrick Linstead is a most valuable contribution to the current discussion of sixth-form specialization. Treating school and university education as a single whole, it comes to two conclusions: firstly that there is little wrong with the balance of studies before the sixth form, and secondly that little can or should be done to broaden the scientific education of university students over the next fifteen years; it is therefore on the sixth form that recommendations for change are concentrated.

The first conclusion is probably true, so far as the actual degree of specialization is concerned, though there seems to be some difference of opinion over the question of premature choice. It is hard to agree, for instance, that by the age of fifteen a pupil should have got sufficient acquaintance with the whole range of subjects to enable him to make an informed choice of such permanent significance. Later statements in the report seem to question this assumption, although only one member specifically wanted to postpone choice between arts and science until the end of the school career. With the second conclusion one can only agree.

The analysis of present-day practice in the sixth form confirms the findings of the Oxford and Birmingham reports to the Gulbenkian Foundation and finds, as they did, that the degree of specialization is too intense. The reforms suggested are very closely in line with the Crowther Report, though slightly more drastic. Like the Crowther Report, the survey is rather vague about how these reforms are to be put into practice, but as a first step they will surely meet with wide, if perhaps ineffectual, approval. A. D. C. PETERSON

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MATHEMATICS

Elements of the Theory of Markov Processes and their Applications, *by A. T. Bharucha-Reid. Pp. xi + 468. McGraw-Hill Book Co. Inc., New York and London.* 1960. 89*s. net.*

This outstanding work is likely to become the standard text on the analysis of Markov processes by methods that do not depend upon measure theory. It effectively presents the detail of many mathematical models, and the main emphasis of the book is on applications. The first three chapters are concerned respectively with processes discrete in space and time, processes discrete in space and continuous in time, and processes continuous in both space and time. The applications are to biology (including diffusion processes in genetics, the theory of epidemics, and radiobiology); physics (particularly cascade processes, particle counters, and nuclear reactors); astronomy and astrophysics (including fluctuations in the brightness of the Milky Way and the theory of the spatial distribution of galaxies); chemistry (including reaction kinetics); and operations research (particularly queueing theory).

From the list of applications mentioned it is evident that no present-day applied mathematician can afford to neglect the study of Markov processes. The same is true of most scientists interested in the theory of their subject. For all such readers the present book offers an excellent introductory account at the graduate level, and the extensive bibliographies are a very good guide to further reading. L. S. GODDARD

Leçons d'algèbre moderne, *by P. Dubreil and M. L. Dubreil-Jacotin. Pp. xi + 393. Dunod, Paris.* 1961. NFcs 35.

This book, which belongs to the series *Collection Universitaire de Mathématiques*, is excellent and will fill a long-felt need for a textbook that introduces the student gently to abstract algebra and at the same time emphasizes those parts that are particularly important in present-day research. It would be most useful if the book were translated into English. The style is not condensed, and many examples are given to illustrate the ideas introduced.

The topics covered are: laws of composition, semi-groups and groups, rings and fields, lattice theory, noetherian rings, vector spaces, algebraic equations, and algebraic field extensions. It is clear that, with this coverage, a detailed treatment of some topics is not possible, and it is a pity that the book contains no references as a guide to further reading. On the other hand, the reader is introduced to a surprising range of ideas: for example, in the chapter on lattice theory, distributive, complemented, and modular lattices are studied, and also the lattice of ideals of a commutative ring. L. S. GODDARD

QUANTUM MECHANICS

Introduction to Quantum Mechanics, *by R. H. Dicke and J. P. Wittke. Pp. xi + 369. Addison-Wesley Publishing Co. Inc., Reading, Mass., U.S.A., and London, W.1.* 1960. 66*s. net.*

This book is intended as an introduction, suitable for students at the graduate level, to the quantum theory. It covers the subject reasonably well and in a fairly condensed form. The standard problems are treated, such as one-dimensional motion, penetration of a barrier, the approach to the classical limit, the properties of angular momentum, the use of matrices, perturbation theory, and scattering theory, and some of these topics are illustrated with the aid of interesting examples. The treatment of quantum statistics at the end is novel for books at this level.

On the whole, the book can be recommended. However, in the reviewer's opinion it does not go sufficiently into the explanation of the physical meaning of the concepts of the quantum theory, which are in fact likely to be extremely strange and puzzling to new students. Also, it would be desirable that, at this level, there should be an introductory treatment of additional topics, such as Dirac's equation, field theory, and some other topics of a similar nature. D. BOHM

DETERGENCY

Surface Activity and Detergency, *edited by K. Durham. Pp. xiii + 250. Macmillan & Co. Ltd, London.* 1961. 35*s. net.*

This volume contains a great deal of information on theoretical and practical aspects of detergency and the relevant parts of surface activity; the wetting of textiles is also fairly thoroughly discussed. The editor himself contributes five chapters giving a great deal of generally accepted theory and many practical considerations. Garrett's chapter on aggregation in detergent solutions is an admirable summary of present scientific knowledge and its historical development. Stevenson's account of the function of foam and of complex formation between detergent solutions and polar substances in the soil calls attention to some less well-known points. Complex formation is further discussed by Lawrence in a brief summary of the work done at Sheffield University, particularly on liquid crystalline formations and myelin figures: he adds many characteristic and pertinent remarks regarding detergency and the vagaries of present nomenclature. Jones's chapter on dirt removal is perhaps a little confusing, but contains a lot of useful information. As a whole, the book is much better balanced than many other multi-author volumes and gives a very good account of the factors involved in detergency. N. K. ADAM

NATURAL FLIGHT

Der Flug der Tiere, *edited by Herta Schmidt. Pp. 164. Verlag Waldemar Kramer, Frankfurt am Main.* 1960. *DM* 8.50.

At the first International Aeronautical Exhibition held in Frankfurt in 1909, considerable interest was aroused by an exhibit of natural flight prepared by the staff of the Senckenberg Natural History Society; the present book is based on an exhibit prepared by the Senckenberg Museum to commemorate the fiftieth anniversary of the original exhibition.

Each of the main types of flying animal is dealt with by an expert, and although specialists may not find much which is not already available elsewhere, there will be a very large number of biologists who will enjoy, and profit from, the clarity of exposition and the very high standard of illustration provided by this book.

To some extent it may be invidious to compare the contributions of individual authors, but by no means the least interesting and scholarly chapter is that by Dr W. Stuve on extinct forms. His text is illustrated by very striking and beautiful photographs of fossil insects, reptiles, birds, and mammals, many of the original fossils being in the Senckenberg Museum. There are also admirable and convincing reconstructions.

Dr Franz deals with living insects, Dr Steinbacher with birds, and Dr Felton with bats and other mammals. These and other chapters necessarily cover relatively familiar ground, but they are all admirably clear, concise, and scholarly. Here again, the illustrations are excellent.

There cannot be the slightest doubt that the exhibition of which this book is the outcome must have been

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of outstanding interest and value. Perhaps some of our older and larger museums might have quite a lot to learn from it.

JAMES GRAY

THERMOPHYSICAL PROPERTIES

Retrieval Guide to Thermophysical Properties Research Literature, *Vol. I, edited by Y. S. Touloukian. Book* 1, *pp.* xii+A1–A11+B–90+1250. *Book* 2, *pp.* vii+169. *Book* 3, *pp.* ix+372. *McGraw-Hill Publishing Co. Inc., New York and London.* 1960. £46 10*s. net.*

The purpose of this publication is to facilitate the location of specific scientific information, thereby simplifying what is becoming an increasingly formidable task.

The Thermal Properties Research Center (TPRC), established at Purdue University in 1957 under Professor Y. S. Touloukian, undertakes the abstraction and dissemination of information regarding fifteen thermophysical properties. This first volume of the Retrieval Guide deals with seven: (1) thermal conductivity, (2) specific heat, (3) viscosity, (4) emissivity, absorptivity, reflectivity (total and spectral), (5) diffusion coefficient, (6) thermal diffusivity, and (7) Prandtl number. It is designed to provide the equivalent of a digital computer for speedily obtaining references to publications relating to these properties.

Three books constitute Volume 1. Book 1 classifies and provides the numerical codes for 14 240 substances; book 2 contains nearly 38 000 entries. It devotes a chapter to each property and, for each code number, indicates the physical state, general scope, language, year, and TPRC serial number; book 3 is the master bibliography and author index for the first 10 000 TPRC serial numbers. The period covered is mainly 1940–59, but some earlier works are included.

The full value of this publication is not to be assessed by conventional means. It emerges when, in response to an urgent enquiry, a list of references can be quickly produced, or the reply, 'no information available', given.

In fields familiar to the reviewer some omissions were noticed, but an occasional new work was located. A few slips have occurred—reference 733 is omitted from the author index, 6204 appears wrongly classified, and a paper (9342) on the thermal conductivity of gaseous chromium seems most unlikely.

A second volume based on searches back to 1920 is promised shortly, and then virtually all the literature on these properties will have been recorded and made available.

Many will be indebted to Professor Touloukian and his colleagues for the lead they have given. Their well-planned work is essential wherever relevant researches or technical applications are in progress.

R. W. POWELL

INFRARED PHYSICS

Infrared Physics, *Vol. I, No.* 1, *March* 1961, *edited by W. K. Weihe, S. Passman, N. Migeotte, and T. S. Moss. Pp.* vi+103. *Pergamon Press, London. Libraries, etc.,* £7; *individuals,* 70*s. net per annum.*

The recent increase of scientific literature makes it important to discourage the establishment of any new journal unless the need for it is clearly recognized. 'Infrared Physics' aims to provide a place for the publications of infrared work relating to space physics, meteorology, astronomy, and geophysics, and to deal specifically with the appropriate instrumentation. The first issue contains useful articles on light sources for communication, detectors, solar spectroscopy, transmitting materials, and other equipment. Whether, in the long run there will be sufficient material of this kind remains to be seen. So long as this journal confines itself to this applied side, it may serve scientific needs. It would be regrettable, however, if it seeks to draw on established media for papers on infrared work relating to physics and chemistry. There is no sharp dividing line in science; measurements on absorption coefficients or band intensities, for example, are just as important in physics and chemistry as in the more applied fields. If this new journal leads to a dispersal of such data it will cause greater inconvenience and cost to libraries and individuals. Otherwise, it may find its proper place.

H. W. THOMPSON

HANDBOOK OF CHEMISTRY AND PHYSICS

Handbook of Chemistry and Physics (*Forty-second edition*), *edited by C. D. Hodgman, R. C. Weast, and S. M. Selby. Pp.* xxv+3481. *The Chemical Rubber Publishing Co., Cleveland, Ohio.* 1961. $12; $12.50 *outside U.S.A.*

This book needs no commendation, as so many laboratories already own well-worn copies of earlier editions, dog-eared in various places according to the owners' interests. A reviewer can but note the changes brought about by the publisher's policy of almost annual revision. This latest edition is extended by a number of new tables: data on diffusion in metals, dissociation constants of organic and inorganic acids and bases, surface tension of organic and inorganic solutes in water and other solvents, lengths of carbon bonds, miscibility tables, and sublimation data for organic compounds. Other tables (atomic weights, electron configurations, analytical reagents and standards, for example) have been revised. When compared with editions only three or four years old, it reflects the trends in science. Thus there is more algebraic information (Fourier transforms and differential equations), and more on space research (atmospheric physics and chemistry, interplanetary orbits), nuclear magnetic resonance, and high-temperature materials. It remains the indispensable laboratory reference work.

J. S. ANDERSON

ORGANIC CHEMISTRY

Cahiers de synthèse organique. Méthodes et tableaux d'application, *by J. Mathieu, A. Allais, and J. Valls. Masson et Cie, Paris. Vol. VI,* Transpositions, *pp.* 417. *Bound, NFcs* 120; *paper covers, NFcs* 110. 1960. *Vols. VII and VIII,* Carbocyclisations, *pp.* 309 *and pp.* 417. *Vol. VII, bound NFcs* 110; *paper covers, NFcs* 100. 1961. *Vol. VIII, bound, NFcs* 85; *paper covers, NFcs* 75. 1961.

These three volumes form part of a series of twelve, of which five have not yet appeared. This work aims at being a general source of organic synthetic methods, and is designed to help the research worker to find and evaluate reactions appropriate to his own particular problem in synthesis. The authors of the three volumes under review achieve this very well indeed, and they are to be congratulated on having produced a work which is both compendious and highly readable.

Volume VI covers rearrangement reactions, such as the Wagner-Meerwein, Wolff, Favorskii, Willgerodt, and allyl rearrangements. Volume VII covers cyclization reactions involving intramolecular carbon to carbon bond-formation, such as the Dieckmann reaction in the homocyclic series and the Pomeranz-Fritsch isoquinoline synthesis in the heterocyclic series (carbon to hetero-atom cyclizations are to be dealt with in a forthcoming volume). Volume VIII covers cyclizations involving carbon to carbon bond-formation

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between two or three molecules (polymolecular cyclizations) such as the Diels-Alder reaction, the Mannich-Robinson synthesis, and trimeric acetylene condensations leading to benzene compounds.

The very large amount of information in these books is well presented. Everything is done in the way of tables and codes to help the reader in finding what he wants. The main body of the work is divided into coded sections, each dealing with a particular type of reaction. The abundant documentation, which includes reviews, covers the literature up to 1959.

These three volumes are of value as works of reference, and can also be read with pleasure and profit as text-books: they should form a valuable addition to any research library. G. F. SMITH

BIOLOGY

Encyclopédie Française, *Vol. IV*, La Vie. Fondement, maintien, reproduction, *Under the direction of P.-P. Grassé. Part* 1, *pp.* x+327+50; *Part* 2, *pp.* iii+329–710+50. *Larousse, Paris.* 1961.

The way of the encyclopaedist must always have been hard; with the proliferation of knowledge it has doubtless become harder, and where biology is in question, perhaps hardest of all. Being an integral part of the universal French encyclopaedia started in 1932, this work does not address itself to the vulgar, but to the élite who wish to form their own opinions on a basis of ordered knowledge; it specifically claims not to save the reader's mind from effort but to stimulate it to activity. The 70 authors who have contributed are, needless to say, all French biologists of distinction, collectively providing a wide coverage.

The individual articles begin with a series on the general characters, origin, and organization of living matter; they proceed to a consideration of the cell, and then to the complex animal, its relations with its environment, and its behaviour. There is a much shorter section on plants and, finally, one on growth and reproduction considered generally. The lower organisms receive only incidental treatment, mainly in the last section. The early chapters are frankly speculative in part, but the middle chapters, so far as the reviewer is competent to judge, are good, up to date compressions of their sometimes unwieldy subjects. There are numerous excellent line illustrations in the text, and equally impressive photogravure reproductions separately bound in each of the two folders into which the volume is divided. There are also an index, a useful bibliography, and an annotated list of contributors.

W. O. JAMES

BIOLOGICAL TECHNIQUES

Section Cutting in Microscopy, *by H. F. Steedman. Pp. vii*+172. *Blackwell Scientific Publications, Oxford.* 1960. 21*s. net.*

This book is a concise but comprehensive text on the embedding and sectioning of biological material for histology. An initial chapter on the history and characteristics of the various embedding media serves to bring some order to their rather confusing variety. The second part of the book includes a theoretical consideration of the properties of these media and the tissues with which they interact, and this helps to supply a rational basis for the methods subsequently described. The third and largest section contains a detailed account of a wide variety of embedding methods and an account of the sectioning of the tissues, including very useful chapters on the difficulties that may be encountered and the care of the microtome knife. It might perhaps have been useful to the novice to give rather more information here on the length of time that the tissues need in the various media. There is a short introductory account of ultra-thin sectioning methods for the electron microscope.

The clarity and order of this excellent book enable it to be referred to with ease; it is especially valuable in its account of new methods, which are based partly on the wide personal experience of the author.

G. N. C. CRAWFORD

BIOCHEMISTRY

Recent Advances in Biochemistry, *fourth edition, by T. W. Goodwin. Pp. vi*+301. *J. & A. Churchill Ltd, London.* 1960. 50*s. net.*

This book, though described as a fourth edition, is in fact a completely new one. The third edition, by J. Pryde, appeared in 1930 and covered entirely different fields. Professor Goodwin has selected eight major topics for this volume. They are electron transport and oxidative phosphorylation, carbon dioxide fixation, phospholipids, regulation of cell metabolism, and the biosynthesis of four classes of cell constituents—aromatic rings, heterocyclic rings, nucleic acids, and terpenes. The aim was to bridge some of the gaps between general textbooks and review articles, especially for the benefit of senior undergraduates, junior research workers, and others who wish to study a comprehensive survey of the selected fields. To such readers the book should indeed be most valuable. Extensive references lead to the original sources. The vast material of the literature is scrutinized with competent judgment and is clearly presented. The chapters are by no means mere compilations of recent advances but are coherent stories, and therefore make most interesting reading. Like its predecessors, the book will no doubt find a large and appreciative readership. H. A. KREBS

Biochemists' Handbook, *edited by C. Long. Pp. xxii*+1192. *E. & F. N. Spon Ltd, London.* 1961. £8 8*s. net.*

This book is a companion volume to the well-known 'Yearbook of Chemistry and Physics' and similar reference books, and contains a vast amount of factual information, some in tabular form, some in the form of brief summaries, on substances of biochemical interest and on biochemical methods. The material has been compiled by 171 experts, mainly from the British Commonwealth and the United States. Subjects covered include the chemical and physical properties of cell constituents and of enzymes, metabolic pathways, and the chemical composition of animal and plant tissues and of microorganisms, as well as more physiological data such as nutritional requirements, chemical composition of foodstuffs, and renal clearance. The sections on methods include sections on manometry, spectrophotometry, chromatography, electrophoresis, and techniques of tissue culture. Of special value are the extensive references to the sources of the information and an index of 8800 entries. The book covers many aspects of biochemistry not dealt with in such similar publications as Rauen's *Biochemisches Taschenbuch* and the 'Scientific Tables' recently published by the Geigy Company of Basle, and it should prove a most valuable time-saver to all who depend on biochemical data. Though primarily designed to meet the needs of research biochemists, it is bound to be of interest to many others.

H. A. KREBS

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Some books received

(*Note.* Mention of a book on this page does not preclude subsequent review.)

GENERAL SCIENCE

Cybernetics (*second edition, by N. Wiener*). *Pp. xvi*+212. *The M.I.T. Press; John Wiley & Sons, New York and London.* 1961. 52*s. net.*

Science and the Future of Mankind, *edited by H. Boyko. Pp. viii* + 380. *Uitgeverij Dr W. Junk, The Hague.* 1961. $9.50.

MATHEMATICS

An Elementary Introduction to the Theory of Probability, *by B. V. Gnedenko and A. Ya. Khinchin* (*translated by W. R. Stahl; edited by J. B. Roberts*). *Pp.* 139. *W. H. Freeman & Co., San Francisco and London.* 1961. 10*s. net.*

Standard Mathematical Tables (*twelfth edition*), *editor in chief: C. D. Hodgman. Pp. ix*+525. *Chemical Rubber Publishing Co., Cleveland, Ohio.* 1960. $4.50

PHYSICS

Applied Optics and Optical Design, *Part 2, by A. E. Conrady. Pp.* 519–841. *Dover Publications Inc., New York.* 1960. $2.95.

A Course in Nuclear Physics, *by D. M. Main. Pp.* 128. *Methuen & Co. Ltd. London.* 1961. 9*s.* 6*d. net.*

General Relativity and Gravitational Waves, *by J. Weber. Pp. viii*+200. *Interscience Publishers Inc., New York; Interscience Publishers Ltd, London.* 1961. *Cloth,* $4.50; *paper,* $2.50.

Growth of Crystals, *Vol. II, edited by A. V. Shubnikov and N. N. Sheftal'.* (*Translated from the Russian*). *Pp. vii*+178. *Consultants Bureau Inc., New York; Chapman and Hall Ltd, London.* 1959. £6 8*s. net.*

Introduction to Elementary Particle Physics, *by R. E. Marshak and E. C. G. Sudarshan. Pp. viii* + 231. *Interscience Publishers Inc., New York; Interscience Publishers Ltd, London.* 1961. *Cloth,* $4.50; *paper,* $2.50.

Optics and Optical Instruments, *by B. K. Johnson. Pp. viii*+224. *Dover Publications Inc., New York.* 1960. $1.65.

Selected Papers on Stress Analysis. *Pp.* 114. *Published on behalf of the Institute of Physics by Chapman and Hall Ltd, London; Reinhold Publishing Corporation, New York.* 1961. 50*s. net.*

CHEMISTRY

Chemistry of Natural Products: Special Lectures presented at the International Symposium, Australia. 1960. (*Reprinted from* Pure and Applied Chemistry, *Vol.* II, *Nos.* 3–4.) *Pp.* 347–635. *International Union of Pure and Applied Chemistry; Butterworths, London.* 1961. 70*s. net.*

Dizionario Tecnico Italiano-Inglese per l'Industria Chimica, *by L. Aghina. Pp.* 431. *Vallecchi Editore, Florence.* 1961. 60*s. net.*

Fatty Acids, their Chemistry, Properties, Production, and Uses, *edited by Klare S. Markley* (*Part 2 of second completely revised and augmented edition*). *Pp. ix*+715–1485. *Interscience Publishers Inc., New York; Interscience Publishers Ltd, London.* 1961. $27.50.

Nitric Acid: Manufacture and Uses, *by F. D. Miles. Pp. ix*+75. *Oxford University Press, London.* 1961. 9*s.* 6*d. net.*

Photochemistry of Air Pollution, *by P. A. Leighton. Pp. ix*+300. *Academic Press, New York and London.* 1961. 88*s. net.*

Progress in Organic Chemistry, *Vol. V, edited by J. W. Cook and W. Carruthers. Pp. viii* + 172. *Butterworths, London.* 1961. 50*s. net.*

GEOGRAPHY

Physical Geography of Asiatic Russia, *by S. P. Suslov* (*translated from the Russian by N. D. Gershevsky and edited by J. E. Williams*). *Pp. xiv*+594. *W. H. Freeman & Co., San Francisco and London.* 1961. £5 5*s. net.*

BIOLOGY

Cells and Cell Structure, *by E. H. Mercer. Pp.* 100. *Hutchinson Educational Ltd, London.* 1961. 12*s.* 6*d. net.*

A Laboratory Manual for Histology (*second edition*), *by J. Forbes. Pp. ix*+132. *Fordham University Press, New York.* 1961. $3.

BOTANY

Mushrooms and Truffles: Botany, Cultivation and Utilization, *by R. Singer. Pp. xxiii*+272. *Leonard Hill* (*Books*) *Ltd, London; Interscience Publishers Inc., New York.* 1961. 60*s. net.*

BIOCHEMISTRY

The Atoms Within Us, *by E. Borek. Pp. xii*+272. *Columbia University Press; Oxford University Press, London.* 1961. 30*s. net.*

MEDICINE

Advances in Blood Grouping, *by A. S. Wiener. Pp. xii*+549. *Grune and Stratton Inc., New York and London.* 1961. $11.

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The Etiology, Pathogenesis and Early Diagnosis of Cancer and Precancerous Diseases of the Stomach, *by K. A. Skulme* (*translated from the Russian*). *Pp.* 441. *Program for Scientific Publications, Jerusalem.* 1961.

A Short History of Clinical Pathology, *by W. D. Foster. Pp. xii*+154. *E. & S. Livingstone Ltd, Edinburgh.* 1961. 27*s.* 6*d. net.*

ENGINEERING

Fortschritte der Hochfrequenztechnik, *Vol. V, edited by M. Strutt and F. Vilbig. Pp. x*+412. *Akademische Verlagsgesellschaft, Frankfurt am Main.* 1960. *DM.*48.

Noise in Electronic Devices. *Pp.* 100. *Published on behalf of the Institute of Physics and the Physical Society by Chapman and Hall Ltd, London; Reinhold Publishing Corporation, New York.* 1961. 35*s. net.*

TECHNOLOGY

Le Bevande Energetiche nel Mondo, *by Lorenzo Vanossi. Pp. vii*+137. *G. B. Paravia & Co., Turin.* 800 *lire.*

Treatment and Disposal of Radioactive Wastes, *by C. B. Amphlett. Pp. viii*+289. *Pergamon Press Ltd, Oxford.* 1961. 70*s. net.*

HISTORY OF SCIENCE

Arbeitserinnerungen, *by H. Staudinger. Pp. xii*+335. *Dr Alfred Hüthig Verlag, Heidelberg.* 1961. *DM.* 28.

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Jean-Baptiste Biot (1774–1862)

Jean-Baptiste Biot was born in Paris on 21st April 1774 and died there just a century ago, on 3rd February 1862. Today he is remembered mainly for his experiments with polarized light, but in his long and active life he contributed not only to mathematics and all the principal fields of physics—heat, light, sound, properties of matter, and electricity and magnetism—but to other branches of learning. His name is commemorated in that of the pleochroic mineral biotite.

Biot's forebears were farmers in Lorraine; his father was employed at the Treasury in Paris. At an early age he found employment in Havre as clerk to a merchant. The monotonous task of copying letters did not appeal to him, however, and he volunteered for the Army as soon as he reached the legally necessary age of eighteen. In 1793 he fought as an artilleryman at Hondeschoote, where the Duke of York, sent to aid the Austrians in Flanders, was decisively beaten and eventually driven back to Hanover. Biot refused the promotion that he might have gained by enlisting as a regular, and sought his discharge. But the mechanism for discharging volunteers was slow, and Biot, young and impatient, set off for Paris without the necessary papers. In doing so, a distinguished scientific career might have ended before it began, for at that time soldiers were forbidden to enter Paris, under pain of death. Biot was arrested and put on trial, but a stranger who had befriended him on the road conducted his defence so ably that he was released. It is said that this friend in need was St Just—the intimate of Robespierre and a leader of the Reign of Terror, who himself went to the guillotine in 1794.

In Paris, Biot studied at what had been the *Ecole des Ponts et Chaussées*, established in 1747 with Jean Perronet (1708–94) as its first director. During the Revolution this was combined with other special schools to form the *Ecole des Travaux Publiques*, which in 1795 became the *Ecole Polytechnique*. On leaving the *Polytechnique* he became for a short time a teacher of mathematics at Beauvais, and during this time he married. In 1800 he was appointed professor of physics at the *Collège de France*; it is said that in the last fifty years of his life he never spent a night away from Paris.

As a young man Biot formed a lasting and significant friendship with Laplace (1749–1827), who was working on his monumental *Mécanique Céleste*, which ultimately appeared in five volumes (1799–1825). Biot begged to be allowed to see part of the manuscript before publication, but Laplace was reluctant; he agreed, however, when Biot made it clear that his desire was to study the book and not to criticize it, and to work through all the calculations. The episode is interesting as giving an indication of Laplace's high opinion of Biot's mathematical ability.

Another episode throws more light on the relationship between the two men. In 1799, Biot sought Laplace's advice on a mathematical paper that he proposed to present to the Academy. The problem was a difficult one, to which Euler had given much attention, but had found only an indirect solution. Biot had made a direct approach and Laplace was enthusiastic about the work, though he pointed out to the young man certain weaknesses in his extension of the theory. Biot abandoned the extension and presented the paper the very next day to an audience that included Bonaparte, who, with Laplace and Lacroix, was appointed an assessor. Bonaparte—of whose value in this context we must be doubtful—appeared with the other members of the committee to approve the paper, even though it met only three days after the *coup d'état* by which he had become First Consul. After the reading of his paper, Biot dined with Laplace. Laplace took him to his study and there showed him a manuscript, presenting an essentially similar treatment of the problem, that he had himself written some time before but had never presented for publication because he had encountered the same difficulties as had Biot in extending the method. Laplace's generosity greatly impressed Biot, twenty-five years his junior; in later life he, too, became known for his kindness to younger scientists.

Biot had a range of interests extending into all the principal scientific fields. In 1804 he developed the theory of heat conduction, checking his calculations by experiment. In the same year he and Joseph Gay-Lussac (1778–1850) accomplished one of the first balloon ascents in the interests of science; they measured the change in the Earth's magnetic field as they ascended, and also made measurements of the density and humidity of the atmosphere. Two years later he went off on another adventurous scientific expedition. This time it was to Spain, whither he accompanied François Arago (1786–1853) to complete the measurement of the arc of the meridian. Their ship was wrecked on the return journey, and Arago narrowly escaped being enslaved at Algiers.

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Biot and Arago collaborated on other work, for example, in making measurements of the density of air and of other gases, and in making an accurate determination of *g* with a carefully worked out pendulum experiment. About 1806, Biot began experiments to determine the speed of sound in solids, and he developed a mathematical theory of its propagation that he published in 1809. With Félix Savart (1791–1841) he investigated the variation of the magnetic field with distance from a long straight wire carrying an electric current, and they formulated the relationship between field and current; this relationship is also known as Laplace's law and Ampère's law. He investigated also the chromatic aberration of lenses, the refractive indices of gases, and the variation of gravitational force with latitude.

But, as was mentioned at the outset, Biot is today remembered mainly for his work on the polarization of light. Huygens had observed the unusual double refraction of Iceland spar, and Newton saw that this was evidence of some sort of asymmetry. The use of the word polarization in this connection derives from Newton's attempts to explain the phenomenon. Biot observed that some liquids, such as solutions of certain sugars, will rotate the plane of polarization; further, he showed the magnitude of the rotation to be proportional to the concentration of the solution. He used his discovery to measure the concentration of such sugar solutions, and applied it in a study of diabetes.

Biot's interest in polarization led to his association with Louis Pasteur (1822–95) in one of the truly dramatic moments in the history of science. In 1844, Biot read to the Academy a note by Bilhardt Mitscherlich (1794–1863), in which he drew attention to the fact that salts of tartaric and paratartaric acid were identical in every respect save that solutions of the former will rotate the plane of polarization of light, while those of the latter have no such effect. This communication greatly interested Pasteur, then one of Biot's students, and he decided to investigate the phenomenon. The consequences are well known, and need not be recounted in detail here. In brief, Pasteur's sharp eyes detected two types of crystal among those of paratartrates, and he was able to separate them by hand. Solutions of one sort of crystal rotated the plane of polarization to the left; those of the other rotated it to the right; when they were mixed in equal quantities there was no effect. Pasteur was, however, then an unknown young man of only twenty-six, and his report was received with scepticism. Eventually, Biot was asked to adjudicate on the young man's claim, and when Pasteur came to make his demonstration he received him coolly. When the striking results convinced him that Pasteur was no impostor, however, his attitude changed entirely. He flung his arms round Pasteur and said: 'My dear boy, my whole life has been devoted to science; what I have just seen makes my old heart beat faster.' Biot himself submitted an account of Pasteur's discovery to the Academy and in later years supported him in many controversies. Pasteur was deeply grateful, and so too was his wife. On 8th December 1862 Pasteur was elected to the Academy, despite intense opposition. On the following morning, outside the gates of the cemetery in Montparnasse where Biot had been buried early in the year, Madame Pasteur waited. When the gates opened, she went, with flowers in her hands, to pay tribute at Biot's grave.

In the course of his own experiments on polarization, Biot observed, in 1812, that the magnitude of the rotation of a beam of polarized light depended on the wavelength of the light used. A century and a half later, this observation has found important applications—under the name of optical rotatory dispersion—in chemical analysis (ENDEAVOUR, **20**, 138, 1961).

Even these extensive investigations in mathematics and science were not the limits of Biot's achievement. He was an accomplished writer, and his textbooks were valued by a generation of physicists. Among them was his four-volume *Traité de physique expérimentale et mathématique* of 1816, a feature of which was its extensive application of calculus to physics; several German editions were published. Another important work, in five volumes, was his *Traité élémentaire d'astronomie physique*.

In 1815 he was elected a Foreign Member of the Royal Society, in the distinguished company of Alexander von Humboldt and Gay-Lussac; he was awarded the Copley Medal in 1840. When, in the fullness of time, the Society published his obituary, particular tribute was paid to his historical researches on Egyptian, Hindu, and Chinese astronomy; his *Précis de l'histoire de l'astronomie chinoise* was an important work. The writer, unfortunately anonymous, considered that in this respect his contributions were comparable to those of many who had worked in no other field. The Royal Society's obituary went on to describe him as the most continuous and varied scientific critic of his time.

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Some geophysical and astronomical aspects of Soviet space research

V. I. KRASSOVSKY

Russian space research policy is based on the belief that although much may be achieved by means of automatic probes, the full conquest of space depends on flights by research vehicles carrying highly trained men. This article describes the principal geophysical and astronomical studies carried out in the Soviet Union as part of the scientific programme in support of the project for manned space flight, first achieved by Yuri Gagarin in April 1961.

Soviet space research began with the launching of numerous geophysical rockets, but the actual breakthrough into space took place in the Soviet Union on 4th October 1957, when the very first artificial satellite was placed in orbit around the Earth. It was followed by other satellites, space probes, and spaceships, some of them carrying biological material. Gradually, the space vehicles were improved and more information was gained about the environment in which they moved, the factors affecting experiments, and the activity of living organisms in outer space. Progress in scientific investigations and space-vehicle design made possible manned flight into space. On 12th April 1961 Yuri Gagarin circled the Earth for an hour and a half on board an orbital spaceship, thus transforming into reality the dream of manned flight outside the Earth. On 6th August 1961, Gherman Titov stayed weightless in space for more than twenty-five hours.

In this article I shall limit myself to a brief review of the main results achieved in the geophysical and astronomical studies carried out in the Soviet Union as part of the scientific programme in support of manned space flight. There have been many, and very important, medical and biological investigations involving the recovery of experimental animals and the use of television equipment, but they lie outside the scope of the present review.

The comprehensive geophysical and astronomical investigations so far conducted may be grouped under the following headings:

1. Evaluation of the hazard, to artificial satellites and spaceships, from micrometeorites.
2. Measurement at different distances from the Earth of hard penetrating and ionizing radiation dangerous to living organisms.
3. Density measurements in the upper atmosphere, related to the drag of artificial satellites.
4. Measurements of the concentration and nature of charged particles in the upper atmosphere and outer space, because these affect radio communication in space.
5. Measurement of the soft components of ionizing radiation in the upper atmosphere. Predominantly this related to corpuscular radiation, as the intensity of hard electromagnetic radiation was known approximately and biological shielding against it presented no difficulties.
6. Studies of the important properties of the Earth and the Moon as celestial bodies. These include the magnetic fields that account for the accumulation of hard penetrating particles; the features of the Moon's reverse side; and many other factors essential to further experiments.

In all these studies, automatic equipment was used; the readings were relayed to the ground by radio. In many cases, the direct relaying of readings was supplemented by an electronic memory that would store the information until a command was given to relay it to Earth, and by instruments that were recovered.

V. I. KRASSOVSKY
Dr. Phys. Math. Sci.
Was born in 1907 in Sevsk (Orel region) and graduated in physics at Leningrad University. From 1930 to 1947 he worked in physical chemistry at a number of research institutes. He was at the Crimean Astrophysics Observatory from 1948 to 1951, introducing astronomical and geophysical researches with electronic telescopes. From 1952 to the present time he has been head of the Department of Physics of the Upper Atmosphere, first at the Geophysics Institute of the U.S.S.R. Academy of Sciences, and then at the Institute of Atmospheric Physics of the Academy. Professor Krassovsky's chief scientific work lies in the field of physics of the upper atmosphere and cosmic space.

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MICROMETEORITES

The impact of micrometeorites on the skin of space vehicles was detected by piezoelectric microphones made of ammonium phosphate. Their signals were fed into a pulse-height analyser for separation according to amplitude and for counting pulses in each amplitude band. The results of measurements [1, 2] are given in the table below. In measuring the mass of micrometeorites their mean velocity was taken to be 40 km sec^{-1}. Referring to the table, it will be seen that the concentration of metoric matter in outer space varies from place to place and with time. It is noteworthy that the concentration was greater at low altitudes and that the mass of micrometeorites in that case was smaller; an accretion of dust particles must therefore exist near the Earth. However, the protection of artificial satellites and spaceships from such micrometeorites poses no particular problems, and meteoritic hazards will not stand in the way of space travel.

PENETRATING RADIATION

A variety of instruments and apparatus was employed for studies of cosmic rays. These include Geiger-Müller counters with various shieldings designed to cut off low-energy particles at a desired energy level; scintillation counters responding to both individual pulses and their total ionization; and Čerenkov counters giving an idea of the composition of cosmic particles.

Sputnik II, launched on 3rd November 1957, had a Geiger-Müller counter with a shielding of 10 g/cm^2 [3–8]. Figure 1 shows the intensity of cosmic radiation as a function of the satellite's position. By far the most striking finding was that the intensity of cosmic radiation grew with altitude. Figure 2 shows the intensity of radiation plotted against altitude above the Earth's surface. The solid curve represents the intensity of cosmic radiation as measured with geophysical rockets, and the dotted line that measured with the aid of Sputnik II. The increase in the intensity of cosmic rays with altitude may partially be explained by the fact that the Earth screens them, and that the particles finding their way into a Geiger-Müller counter at high altitudes will come from a larger region than at lower altitudes. This explanation, however, can account only for an increase of 10–15 per cent at an altitude of 700 km. The observed increase is considerably greater than this, pointing

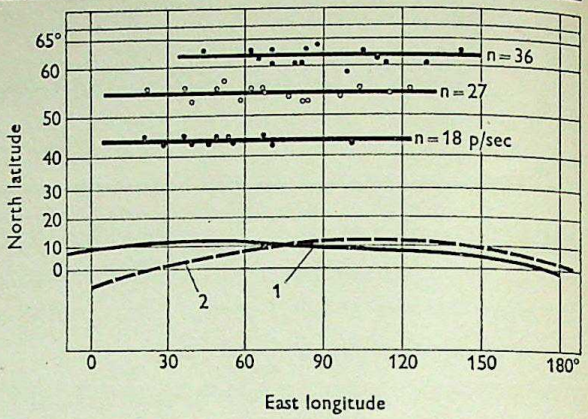

FIGURE 1 – *Intensity of cosmic rays plotted against latitude and longitude. The intensity (n) is the number of pulses per second registered by a Geiger-Müller counter. (1) is the equator of cosmic rays according to Stimson and (2) the equator of the geomagnetic dipole.*

Vehicle	Date	Mass of particles observed (g)	Number of impacts per m^2 per sec
Sputnik III	15.5.1958 16–17.5.1958 18–26.5.1958	8×10^{-9}–$2{\cdot}7\times10^{-8}$	4×10^{-4} 5×10^{-4} 10^{-4}
First space probe	2.1.1959	$2{\cdot}5\times10^{-9}$–$1{\cdot}5\times10^{-8}$ $1{\cdot}5\times10^{-8}$–2×10^{-7} 2×10^{-7}	2×10^{-3} 5×10^{-4} 10^{-4}
Second space probe	12.9.1959	2×10^{-9}–6×10^{-9} 6×10^{-9}–$1{\cdot}5\times10^{-9}$ $1{\cdot}5\times10^{-8}$	5×10^{-5} 5×10^{-5} 9×10^{-5}
Third space probe	4.10.1959 18.10.1959	10^{-9}–3×10^{-9} 3×10^{-9}–8×10^{-9} 8×10^{-9}	4×10^{-4} 2×10^{-3} 4×10^{-4}

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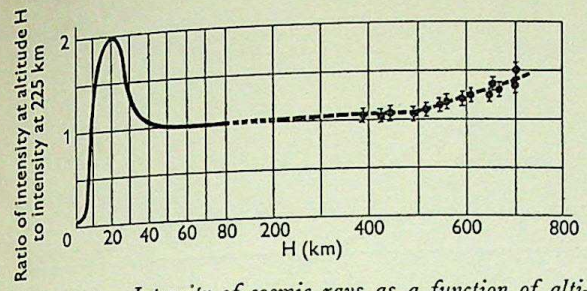

FIGURE 2 – *Intensity of cosmic rays as a function of altitude(H), taking the value at 225 km as unity. Limits of error are indicated by horizontal lines above and below the points indicating the values measured.*

unambiguously to other factors. Somewhere between 04.30 and 04.41 hrs. Moscow time on 7th November 1957, Sputnik II indicated an increase of up to about 50 per cent above normal average intensity at latitudes above 58° North. Figure 3 shows the increase plotted against time. Further experiments confirmed an increase in the intensity of hard radiation in space near the Earth, and a decrease to the normal level at a distance of several Earth radii.

The terrestrial magnetic field was found to cause accumulation of a huge quantity of energetic particles, the most penetrating being concentrated in the inner zone at an altitude of 1000 km above the Earth's surface (the inner radiation belt). An extensive external radiation belt was also found, containing less penetrating particles. Figure 4 gives the results obtained by means of the first Soviet space probe, fired on 2nd January 1959. A large number of observations seem to prove that the intensity of penetrating radiation varies with time, and that its distribution in space near the Earth does not remain constant.

In other experiments [9–11], Čerenkov counters identified particles with charges both equal to and greater than unity. In some cases, an increase in the number of heavy nuclei registered was observed during solar flares [41], but their relative number was small. In addition, an increased leakage of hard particles was noted in the areas of geomagnetic anomalies [42]. Cosmic rays were also studied by means of stacks of photographic emulsion that could be recovered after exposure.

Such measurements of the increase in the intensity of highly energetic particles served as a guide for the development of biological shielding for living creatures sent into outer space. In passing, it may be remarked that the penetrating particles in the inner radiation belt were found to be the more dangerous.

DENSITY OF THE UPPER ATMOSPHERE

Before Sputnik I was launched there was no reliable information about the density of the atmosphere at the altitudes above 200 km in which it was intended that artificial satellites would travel. The only estimate that could be made was based on extrapolation of observations made by means of rockets in the lower atmosphere. A hypothesis was advanced that above 200 km the atmosphere was isothermal, had a low density, and a temperature of not more than 1000° K. As the atmosphere exerts a drag on satellites, measurement of the drag makes possible a direct estimate of atmospheric density. As both the technique and the details of the calculations may be found in the literature [12–15], I shall here limit myself to the discussion of the final results.

The quantity determined from observations of the change in period of the satellite's orbit is approximately $\rho\sqrt{h}$, where ρ is the density of the atmosphere near the perigee of an Earth satellite, and h is the 'scale height' in the same region. For the precise value of ρ to be determined, assumptions have to be made about the scale height and, consequently, about the average molecular weight, temperature, and temperature gradient of the atmosphere in that region. On all reasonable assumptions, the error in the value of h could not appreciably mask the true value of the average density at 220 km, which was the perigee of the first sputniks. Supposing that the scale height rises from 30 km at about 200 km altitude to 60 km at about 400 km altitude, the density is of the order of $2{\cdot}5 \times 10^{-14}$ g/cm^3. This is appreciably more than had been expected before the first satellites were launched. In the daytime, the density

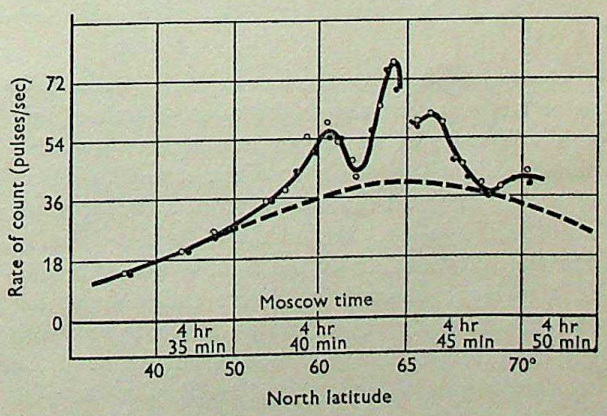

FIGURE 3 – *Changes in the intensity of cosmic rays during a sporadic flare. The broken line indicates the average value of the intensity under normal conditions; the solid curve shows the variation in intensity during the flare.*

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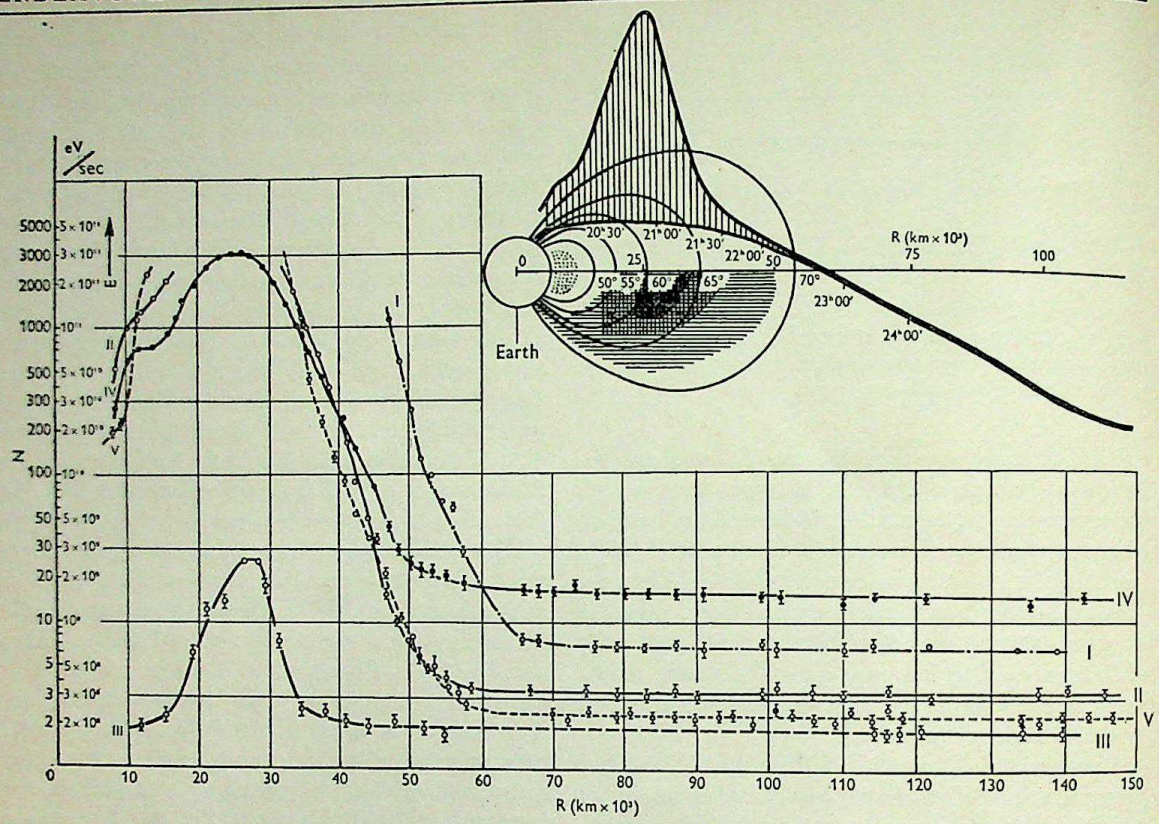

FIGURE 4 – *The trajectory of a rocket, in geomagnetic coordinates, is shown in the top right corner. Time of flight (Moscow time) is indicated along the trajectory, while the intensity of penetrating radiation is indicated by the vertical lines running to the trajectory; R is the distance from the Earth's centre. A measure of intensity is provided by total ionization (curve IV). The figure shows magnetic lines of force intersecting the Earth's surface at geomagnetic latitudes 50°, 55°, 60°, 65°, and 70°. The magnetic field is assumed to be the field of a dipole with a geomagnetic pole at 78·5° North latitude and 69° West longitude. Dots indicate the inner radiation belt and lines the outer radiation belt. The shading gives a qualitative idea of the distribution of intensity in the outer radiation belt.*

The curves show the intensity as a function of altitude. Curves I, II, and III indicate the rate of count (N) for particles with a threshold of 45 keV, 450 keV, and 4·5 MeV respectively; the rate of count refers to unit cross-sectional area of the crystal (19 cm²) used. Curve IV shows total ionization, that is total energy given up by the scintillating crystal per second. Curve V gives readings of Geiger-Müller counters; the number of pulses is referred to unit cross-sectional area of the counters (4 and 15 cm² respectively). Readings of the two counters agreed reasonably well; in the case of high intensities, readings of the smaller counter only were used.

of the atmosphere at 220 km in high latitudes is some ten per cent above the average value [12].

The density of the upper atmosphere was also calculated [16] on the basis of the rate of diffusion of a sodium cloud discharged from a rocket at an altitude of 430 km. At that time, the cloud was in a region illuminated by the Sun, which caused it to fluoresce; observations were made from the Earth, where it was still dark. The rate at which the cloud expanded gave a direct measure of the concentration of atoms and molecules at that altitude; the value found was $2{\cdot}3 \times 10^{-16}$g/cm³.

The sputniks were useful also in making pressure measurements by means of manometers [17]. As distinct from a rocket rising vertically to an altitude of several hundred kilometres within a few minutes, an artificial satellite stays in the upper atmosphere for a long time. Any gas that may have been adsorbed by the satellite completely volatilizes after a few circuits of the Earth; consequently gas given up by the equipment cannot falsify the values. The number of particles (atoms and molecules) per cubic centimetre, in the upper atmosphere, as measured by a variety of techniques, is summarized in figure 5.

IONIZATION

Before artificial satellites and high-altitude rockets were launched, there had been no direct information about the ionization of the atmosphere

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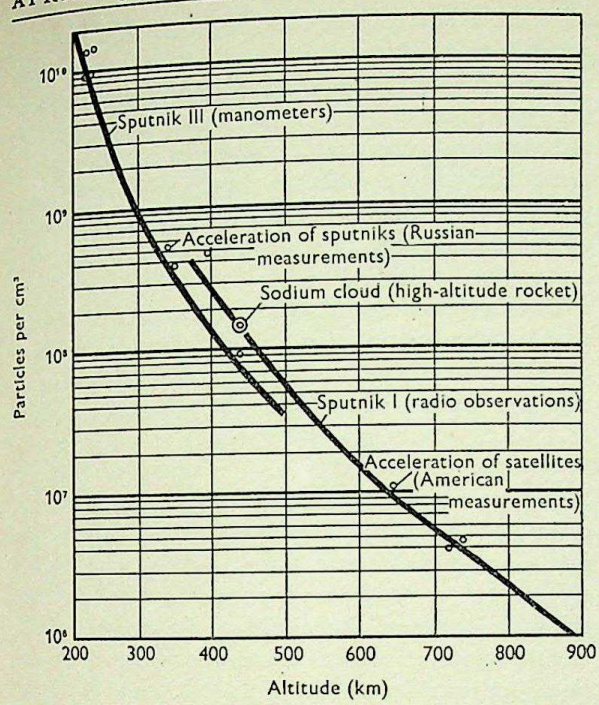

FIGURE 5 – *Variation with height of concentration of atoms and molecules in the upper atmosphere.*

above the F-region of the ionosphere. As a start, the concentration of electrons in the upper atmosphere was determined by means of an ultra-high-frequency dispersion interferometer lifted by rockets. Variations in the concentration of electrons with altitude were determined by observing the phase differences of two coherent radio waves of different wavelengths which were either multiples or submultiples of each other [18–21]. Quite a number of measurements were made by this method, covering the distribution of electron concentration both below and above the maximum of ionization, which occurs at about 300 km; figure 6 summarizes some of the measurements. It was established that the concentration of electrons increased almost steadily with altitude to a maximum, and then dropped. The total number of electrons above the maximum of ionization was larger than that below it.

Later, various designs of charged-particle traps mounted on artificial satellites and space probes were used [18–27]. In the traps was a number of grids placed in front of the collector and kept at different potentials relative to the vehicle's hull or the instrument container. With this device it was possible to measure the density of positive-ion streams at various predetermined energy levels, both in the upper atmosphere and in outer space. Figure 7 shows a typical distribution of ion densities up to an altitude of 750 km, as determined by means of ion traps. The density of ions was found to be still of the order of several hundred ions per cm^3 up to 15 000 km from the Earth's centre, but dropped at greater distances. At distances of over 20 000 km it was possible to determine only the upper limit of ion concentration, which was less than ten ions per cubic centimetre. Under normal conditions in outer space, the measured density of ions was never found to exceed several ions per cubic centimetre. During the magnetic storm of 17th February 1961, however, the Soviet Venus probe traversed a stream of ions with a flux of up to 10^9 ions per cm^2 per sec. In this case, the cause was undoubtedly a corpuscular stream emitted by the Sun.

The charged-particle traps mounted on space probes have established that at some distances from the Earth the flux of electrons of certain energies colliding with the probe is greater than the total flux of positive ions impinging upon it.

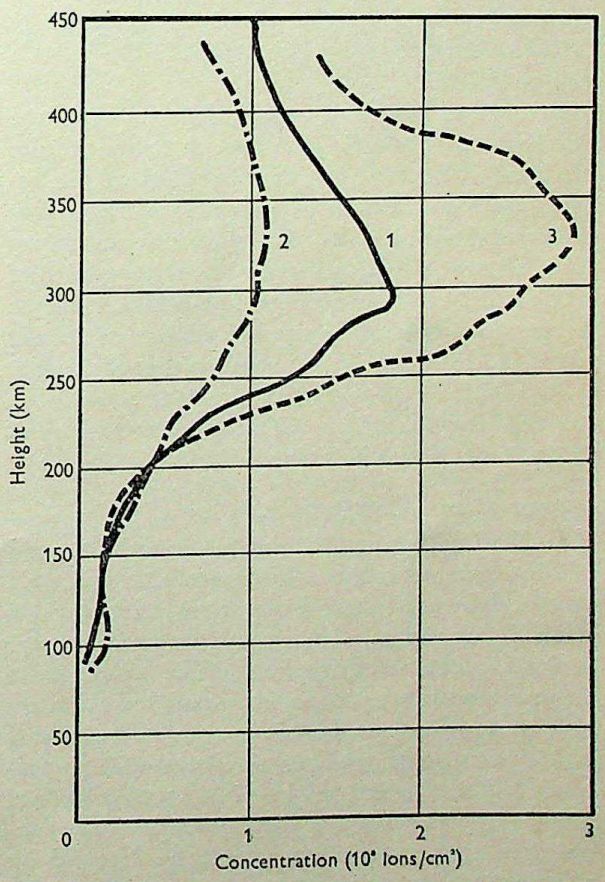

FIGURE 6 – *Electron concentration as a function of altitude. Curve I,* 11.40 *hrs,* 21*st February* 1958*; Curve II,* 08.06 *hrs,* 27*th August* 1958*; Curve III,* 15.54 *hrs,* 31*st October* 1958.

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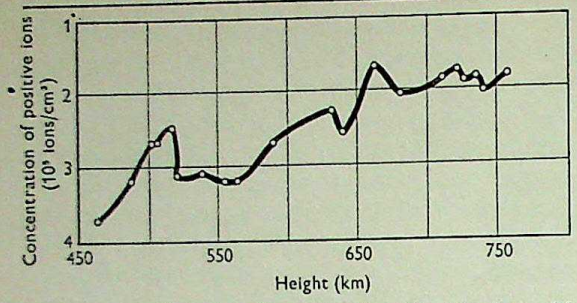

FIGURE 7 – *Variation of concentration of positive ions with altitude at* 17.00 *hrs*, 15*th May* 1958.

This indicates a change in the distribution of energy between ions and electrons in space. At distances up to 50 000 km the flux of electrons with an energy higher than 200 eV did not exceed 3×10^7 per cm^2 per sec. At distances from 50 000 to 75 000 km, however, it was as high as 2×10^8 per cm^2 per sec. This seems to indicate the existence of a zone where ionized interplanetary gas penetrates into, and moves on, the periphery of the terrestrial magnetic field. This zone is situated beyond the region where charged particles of higher energies are concentrated in the Earth's magnetic field.

In another group of experiments [28–32], the composition of atmospheric ions up to an altitude of about 1000 km was studied by means of a mass-spectrometer mounted on geophysical rockets or on an artificial satellite. The predominant ion up to an altitude of 250 km was found to be the ion of nitric oxide, while at higher altitudes, atomic oxygen ions prevailed; the abundance of atomic nitrogen ions was only a few tenths that of oxygen.

A wealth of data on the ionosphere has been derived from analysis of radiocommunication between the Earth and the sputniks [33].

GEOMAGNETIC CORPUSCLES

In May 1958, an effort was made, with the aid of the equipment borne by Sputnik III [34, 35], to detect in the upper atmosphere electrons responsible for auroral displays and for an additional ionization of the atmosphere. The existence of these electrons was to be indicated by two very thin phosphors (2 mg of silver-activated zinc sulphide per square centimetre) coated with aluminium foil of different thickness (0·4 and 0·8 mg of aluminium per square centimetre). The fluorescence of the phosphors was to be detected by photomultiplier tubes coupled to an electronic circuit for amplification. The resultant signal was to be fed into a memory device and relayed to the ground on a command from the Earth. Provision of metal foils of differing thickness made it possible to assess both the intensity and the energy of the fluxes of electrons that made the phosphors fluoresce most brightly. An important feature of the apparatus was that for all practical purposes it responded only to electrons, ignoring protons and photons even if their energy flux was many orders of magnitude higher than that of the electrons under observation.

In this experiment, powerful fluxes of electrons, with an energy of about 10 keV, were observed at an altitude of up to 1900 km in high-latitude regions. The intensity of the fluxes was very high, and in most cases was beyond the range of the apparatus, as fluxes of such magnitude had not been expected. Throughout the experiment the effective energy of electrons varied, while the intensity of the fluxes tended to drop as the effective energy of the electrons rose. In the 'beyond-the-scale' state of the counter, the energy of the electrons under observation was upward of 100 ergs per cm^2 per sec at an altitude of about 1900 km. For the most part, the registered electrons were moving approximately in a plane normal to the magnetic lines of force, and there were more electrons moving along the magnetic lines of force towards the Earth than in the reverse direction. The electrons moving towards the Earth at a small angle to the magnetic lines of force, that is, having a magnetic moment enabling them to penetrate into the atmosphere as low as 100 km, might contribute an energy as high as 1 erg per cm^2 per sec.

This experiment yielded for the first time some information about the existence at high altitudes (up to 1900 km) of high concentrations of electrons with an energy predominantly about 10 keV. All other investigations of such electrons, both before and after this experiment, relate to appreciably lower altitudes. No particularly intense flux of similar electrons had been detected in previous experiments. If the electrons observed in the experiment described above could reach the lower regions of the atmosphere, they would noticeably increase its ionization and heating; in fact, this does occur sporadically.

Another type of equipment [4], intended for studies of cosmic rays and also mounted on board Sputnik III, detected electrons of somewhat higher energy concurrently with the experiment just described. When the sputnik was traversing high-latitude regions, the heavily shielded scintillation counters had to operate beyond their normal range. This seems to have been due to X-rays generated

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by the bombardment of the hull by powerful fluxes of electrons with an energy of over 20 keV.

As compared with the highly penetrating particles described earlier in this article, electrons with an energy of less than 100 keV do not constitute any appreciable danger to astronauts. The X-rays generated by electrons with an energy varying from 20 to 100 keV can be attenuated to tolerable doses by a relatively thin shield. These electrons, however, extend down to below 100 km during geomagnetic disturbances, and increase the ionization of the denser atmosphere to such an extent that it will strongly absorb radio waves.

STUDIES OF CELESTIAL BODIES

In a number of experiments [36–39] the intensity of the terrestrial magnetic field at different distances from the Earth was measured. These measurements were made necessary by studies of the corpuscular radiation retained by the terrestrial magnetic field. Figure 8 shows the results of measurements made at different times. The differences in the variation of the strength of the magnetic field may be explained both by disturbances in its spatial distribution and by its variation with time. The flight of space probes lasts longer than the period within which normal fluctuations of the terrestrial magnetic field are to be expected.

Of interest also are studies of the Moon [40], the reverse side of which was photographed by means of an automatic space probe. The probe was automatically brought into such a position that pictures of the Moon's hidden side might be taken. The film was then developed, and the image was relayed to the ground by radio-telegraph.

Somewhat earlier [39], the magnetometer mounted on a lunar probe had established that the magnetic field near the surface of the Moon did not exceed 10^{-3} gauss. The absence of an appreciable magnetic field round the Moon was confirmed by the fact that the devices intended for investigations of cosmic rays [7] failed to indicate any increase in the intensity of penetrating particles near the Moon compared with their intensity in outer space. If the Moon had a magnetic field it would inevitably accumulate such penetrating particles, arising from the decay of neutrons ejected from the Moon's surface through irradiation by primary cosmic rays.

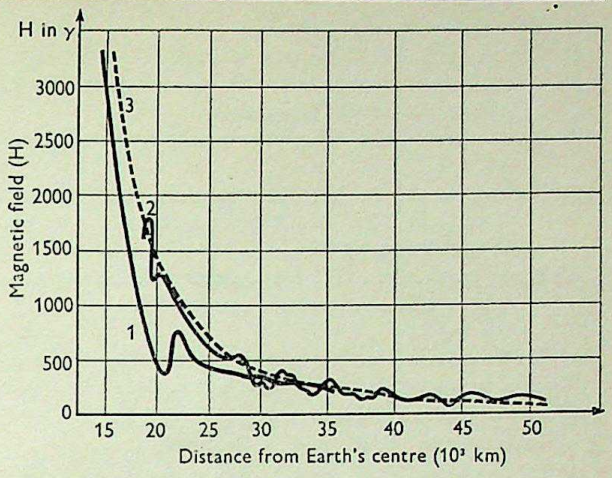

FIGURE 8 – *Intensity of the magnetic field (H) as a function of distance from the Earth's centre. Curve 1 is based on data obtained during the flight of the first space probe, launched on 2nd January 1959; Curve 2, on data obtained during the flight of the second space probe, launched on 12th September 1959; Curve 3 shows changes in the ideal intensity of the magnetic field as affected by dipole and quadrupole terms.*

CONCLUSION

The geophysical and astronomical investigations carried out in the Soviet Union by means of artificial earth satellites, space probes, and geophysical rockets have yielded information essential for the design of manned satellites and spaceships. At the same time this information is of independent interest from the viewpoint of pure science, as it has led to the discovery of new phenomena—such as the accumulation of penetrating particles in the terrestrial magnetic field—and to the more accurate determination of many quantities that were not precisely known before, such as the density and ionization of the upper atmosphere. This wealth of new information is currently exciting much lively discussion and speculation. A large number of stimulating hypotheses have been advanced, for example, in explanation of the origin of penetrating radiation in the terrestrial magnetic field. The future holds out promise of still more successful investigations with still better equipment.

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The synthesis and uses of artificial gemstones

E. A. D. WHITE

Attempts to make gemstones have been fully successful only relatively recently, but it is now possible to make stones that are more pure than those that occur naturally. These are important because of the light they throw on the nature of the crystalline state, and because of their application in 'low-noise' amplifiers, masers and lasers. This article deals with the methods of synthesizing gemstones, and, in particular, with the newer developments in technique and the improvements in the crystalline perfection of the products.

The history of attempts to make gemstones is a long one, but little success was achieved until the beginning of the twentieth century—rubies were first synthesized in 1904. It was at first possible to produce only a few types of synthetic stone, and those on only a limited scale. Not until after the last war, when improved refractory materials and more powerful apparatus made it possible to use more extreme conditions, was any marked progress made. More recently, developments in solid-state physics have led to a revival of interest in gemstone synthesis. It has become increasingly important to be able to study physical processes in single crystals, and those grown artificially have been extensively used for research; they have also found industrial application. The particular interest of gemstones lies, firstly, in their hardness and durability, and secondly, in the changes in physical properties, that, with the colours of the stones, are caused by trace impurities.

Hard materials, in which the atoms are strongly bound to one another, are particularly important industrially for cutting and abrasion, for wear-resistant surfaces, and as chemically inert refractories and structural materials. A considerable part of materials research is at present devoted to the study of such materials and the search for new ones with improved properties.

Many of the earlier techniques of synthesis resembled natural processes, although it is not usually possible to reproduce in the laboratory the duration, quantities, or pressures involved. However, the natural formation of quartz crystals from localized hot igneous intrusions is closely paralleled by hydrothermal syntheses, and crystals formed from a fused melt often closely resemble those from a cooled natural magma.

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Was born in Kent in 1926 and graduated at Brighton Technical College. He undertook research at Aberdeen University in the field of hydrothermal synthesis, and later entered industry. His present activities cover a broad interest in all aspects of crystal growing and the preparation and study of inorganic materials for use in research.

SYNTHESIS UNDER ULTRA-HIGH PRESSURE

The pioneers in the field of ultra-high pressures used low temperatures, in general, because the materials used in the equipment quickly lost their strength when heated. The problem of simultaneously achieving both high pressures and high temperatures was solved by the adoption of internal heating; massive supports that allowed the heat generated to be quickly dissipated made it possible to retain the physical strength of the containers. Part of the success was due to the use of pyrophyllite (akin to talc) as a gasket material and pressure-transmitting medium; it has the advantage of a considerable increase in melting point with increasing pressure. The first recorded use of internal heating, in this form, at ultra-high pressures was in the synthesis of diamond by the General Electric Company of America [1], although several other laboratories were evidently engaged on similar projects. H. Tracy Hall, in particular pioneered much of this high-pressure work, and he was associated with the development of several novel types of apparatus [2].

The most successful of the diamond-forming devices consists essentially of opposing pistons that compress a small region within a specially shaped bolster (the 'belt'), constructed from a number of preshrunk rings of high-strength steel (figure 1). To obtain adequate sealing and pressure transmission, a complex array of suitably shaped pyrophyllite and metal gaskets are used that fit between the pistons and the belt, and closely fit

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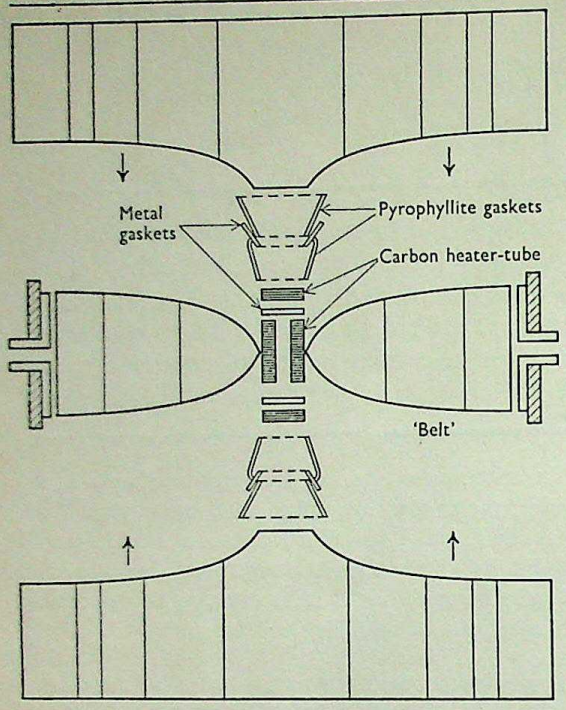

FIGURE 1 – *Ultra-high-pressure apparatus. In use, the nest of gaskets slides over the carbon heater-tube, which contains the reaction material, so as to fit against the 'belt'.*

round the central cylindrical carbon heater-tube. The latter encloses the working space and provides resistance heating by electricity conducted through the pistons. The current version of this equipment can reach pressures of 110 000 atmospheres at 3500° C and maintain these conditions for an indefinite period. Even higher pressures, of 160 000 atmospheres, can be reached in modifications of the apparatus, and higher temperatures, too, can be attained for short periods.

Some difficulty is experienced in accurately measuring the pressures and temperatures attained in the various forms of apparatus. This is due to some uncertainty about the effect of pressure on the thermocouples used, and about the influence of temperature on the physical transitions in various substances which are used as pressure calibration points. It is unlikely, however, that the estimated values are far wrong, and there is no doubt that such extreme conditions of temperature and pressure have not previously been obtained simultaneously.

The belt type of apparatus requires a high-capacity hydraulic press for its operation and is not simple to construct. The tetrahedral-ram equipment, which was devised at about the same time, offered the possibility of achieving ultra-high pressures cheaply and with commercially available equipment. Using four 500-ton hydraulic rams with tungsten carbide anvils, mounted on a tetrahedral frame and converging on the centre, a machine with a working space of about 15 cm³ can provide pressures up to 80 000 atmospheres. This design has been largely superseded by an elegant modification in which only a single hydraulic ram is required [3], the force in the other directions being obtained by reaction of the anvils bearing on an internal conical surface (figure 2). In order to allow for slight movement of the anvils and to provide electrical insulation, thin sheets of Teflon are inserted under each anvil.

The latest development is a metal sphere cut into four suitably shaped segments, with tungsten carbide anvils inserted in them. This sphere is surrounded by a rubber shell and fits into a chamber in which the pressure is produced by a conventional high-pressure hydraulic pump. The pressure obtainable depends on the ratio of the radius of the sphere to that of the internal working space. This design has the advantage that no large or elaborate rams are required; the whole apparatus can be contained in a small space.

In all types of anvil apparatus, the central tetrahedral spaces are packed with specially shaped pyrophyllite pieces allowing a carbon cylinder to be inserted and heated by electricity conducted through two of the anvils. The materials to be studied are packed in the centre of the heater tube. Alternative configurations of the working spaces are possible, although the increased volume obtained is offset by the increase in complexity due to extra anvils, and the slightly decreased

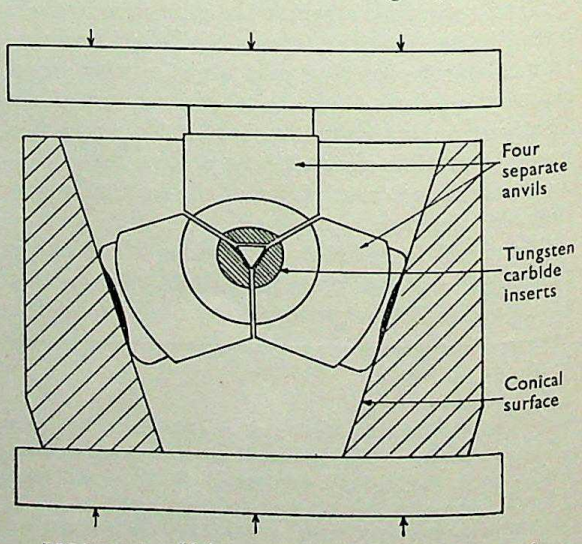

FIGURE 2 – *Uni-ram, tetrahedral anvil, equipment.*

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strength of the anvils due to modification of their shape.

The possibility of working at combinations of high pressure and high temperature has opened up many possibilities for the preparation and study of materials. Apart from diamond, borazon (the cubic modification of boron nitride) has been synthesized, as has a type of garnet [4]. Studies of the physical properties of materials under very high pressures have already been started in several laboratories.

Successes by other techniques for diamond synthesis have recently been claimed, but no information has so far been released. A Dutch process is claimed to produce diamonds under dynamic high-pressure conditions that are probably obtained by the explosion of a shaped charge, but a method of this type is not likely to yield crystals of useful size.

FLAME FUSION

The original equipment designed by M. A. Verneuil [5] is still used, in virtually unchanged form, for the production of gem-quality ruby crystals. In this process, a free-flowing alumina powder is contained in a hopper over a fine-mesh gauze; regular tapping of the container causes trickles of powder to fall into the central oxygen feed-pipe to the burner. In passing through the flame the powder is melted and falls on to the molten top of the growing crystal, which can be withdrawn slowly from the flame as it grows.

To start, a small pyramid of unfused powder is built up at a reduced flame-temperature. As the flame becomes hotter, the tip of the cone melts and growth proceeds from this point. In most cases a single crystal develops, as secondary crystals that are not favourably aligned tend to stop growing after a short time. The crystallographic orientation of the resulting 'boule' is random, and it is preferable to start growth on a small length of seed crystal, as this gives a more certain result and allows crystals of any desired orientation to be grown. Suitable seed-rods can be grown in a smaller version of the furnace fitted with an automatic photoelectric device for lowering the crystal. Single-crystal rods up to 45 cm long can easily be obtained in this way.

Although the method is simple in concept, the factors affecting growth by flame fusion are many and complex. Efforts have been made to make the apparatus automatic in order to be less dependent on the skill of the operator and to improve the quality of the products. There is an improved

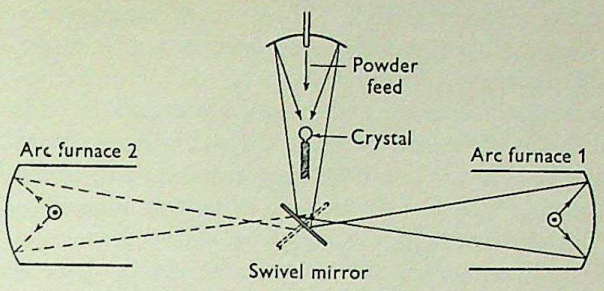

FIGURE 3 – *Arc-image furnace.*

version of the furnace, in which a vibrating feed-system gives a controlled even feed of powder, and the temperature of the furnace is regulated by a radiation pyrometer that controls the oxygen supply. Provision can also be made to mechanize the critical initial stages of growth until the crystal has attained the size desired.

The crystals produced are in a highly strained condition because of the temperature gradients in the furnace during growth and the rapid cooling when the flame is shut off. Spontaneous cracking is frequently encountered, and the crystals must be annealed by heating at a high temperature before they can be cut or ground.

The Linde Air Corporation has attempted to overcome some of these problems by rapid rotation of a crystal seed-rod held horizontally, so that the perimeter of the growing disc is formed under virtually isothermal conditions. A ruby boule grown by this technique is shown in figure 13.

The flame-fusion process has recently been extended in scope in two directions. Firstly, by the use of volatile halides as the feed material, which enables a much closer control of the rate of growth to be obtained and also makes it possible to purify the raw material by distillation, and secondly, by the use of heat sources other than a flame. In this class are the arc-image (figure 3) and plasma-jet furnaces adapted for Verneuil-type growth. Both processes allow control of the growth atmosphere and consequently can be used for materials that would be decomposed in a flame, such as nitrides, carbides, and silicides. The plasma-jet is coming into general use as a source of high temperature for cutting and flame-spraying, but here the usual blown jet is of restricted use because of difficulties in injection of the feed material. A more useful type of 'burner' enclosing a plasma generated by electrical induction has been used successfully for growing alumina and zirconia crystals and should prove capable of melting magnesia [6].

The flame-fusion process in general suffers from the inherent disadvantage of an uneven

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temperature pattern. This causes inhomogeneity when volatile impurities are incorporated. Further, the amounts of such impurities that can be incorporated are limited, and in some cases it is difficult to ensure that the desired valency states are retained. The following methods are much more suitable for the growth of 'doped' crystals—those grown with deliberately added impurities—and are potentially capable of giving crystals of much higher quality than the two processes described above.

HYDROTHERMAL SYNTHESES

Although many compounds are insoluble or relatively insoluble in water at temperatures up to 100° C, at higher temperatures the solubility increases considerably. It may become sufficient to make it possible to obtain small crystals from a solution. Experiments have shown that there is no discontinuity in solubility/temperature curves at the critical point, and crystallization occurs equally well from the 'fluid' state. Rates of crystallization are slow compared with processes using higher temperatures, but the quality of the crystals obtained is generally higher, and may in many respects be higher than that of natural crystals of the same species [7].

The hydrothermal process has met with most success in the growing of quartz and other siliceous materials. These are sufficiently soluble in aqueous alkali to be grown at moderate temperatures, about 400° C. It is difficult to obtain crystals of these materials by other techniques because the melts are of high viscosity and tend to form glasses.

The simplest, and probably the most successful, type of equipment used for quartz production is shown in figure 4. As the aqueous solution must be contained at high temperatures, and consequently high pressures, it is necessary to use a thick-walled vessel or autoclave. This contains the nutrient material, the aqueous solvent, and seed-crystal plates suspended from the lid. It is heated by a hotplate. This produces a temperature gradient along the length of the vessel. The nutrient material at the base dissolves and the solution rises by convection; in the cooler region at the top, the solution becomes supersaturated and growth on the seed crystals occurs. The cooled solution then returns to the bottom. The temperature gradient, and hence the rate of growth, can be modified by using an internal baffle to reduce circulation. The pressure attained is regulated by the quantity of solution added initially, and the

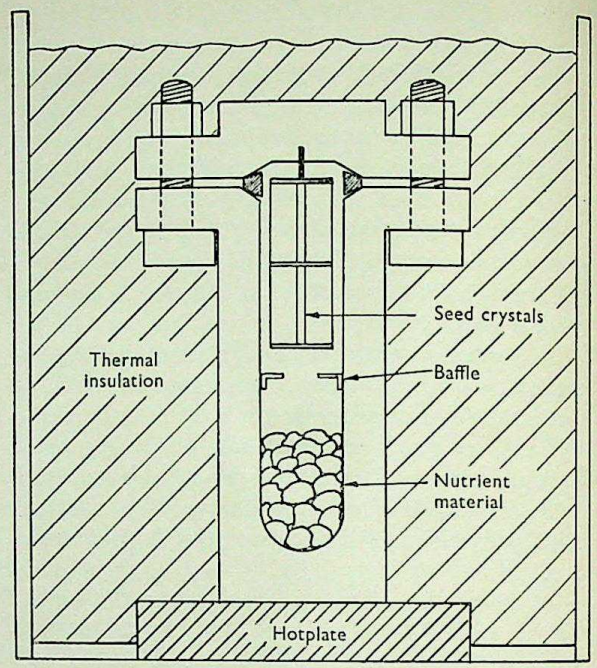

FIGURE 4 – *Temperature-gradient hydrothermal apparatus.*

temperature is controlled by a thermostat. The solubilities of many siliceous materials are higher in alkaline solutions than in pure water, and sodium carbonate or sodium hydroxide solutions are commonly used [8].

The autoclaves used for the growth of quartz are constructed from mild steel, which can be used at pressures of 1000 atmospheres and temperatures of 450° C. Less soluble materials, or those that are stable only at higher temperatures, necessitate the use of creep-resistant alloys; thus corundum crystals are grown in stainless steel autoclaves at temperatures up to 650° C [9]. Above 500° C, the autoclaves are markedly attacked by water, and the crystal products are therefore contaminated with iron. It is therefore desirable to use a platinum liner both to preserve the autoclaves and to obtain a pure product.

The limitations of the hydrothermal method are the cost of the equipment; the impossibility of observing growth or controlling the processes other than by external control of temperature; and the slow rates of growth.

FLUXED-MELT GROWTH

The fluxed-melt technique provides an alternative method that is reasonably fast, can be applied to many different materials, and uses readily available equipment. It is similar in many respects to crystallization from aqueous solutions,

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but the solvents used are solids with relatively high melting-points. The alkali halides and carbonates, lead oxide, and lead fluoride are of particular interest for this work.

The material to be grown, or its constituents, is placed in a platinum crucible with sufficient solvent to form a homogeneous solution on heating. A conventional electrical resistance furnace is used, preferably one that can be controlled in temperature and cooled over a period of hours or days. Since many of the fluxes used are volatile at about 1000° C, it is desirable to seal the crucible; otherwise excessive evaporation, with the production of a large number of small crystals, occurs. As the melt is cooled, the solution becomes supersaturated, and if cooling is slow, only a few crystal nuclei will develop. Slow cooling is continued until the melt solidifies, or, alternatively, until the solvent can be poured off just before solidification.

The main disadvantage of the technique is the difficulty in controlling nucleation; this is aggravated by the difficulty in preventing loss of solvent by evaporation. A novel solution to this problem was achieved by J. W. Neilsen [10], who grew yttrium iron garnet ($Y_3Fe_5O_{12}$) crystals from a melt of a composition such that another crystal species, which utilized many of the nucleating sites normally available for growth, was initially formed. The formation of this species caused a gradual change in the composition of the melt, and a slow approach to the condition for crystallizing the garnet was possible. Crystals grown by this technique represented an improvement over previous attempts at the growth of ferrite materials, but even better results are obtained by the use of a low-viscosity flux, a mixture of lead oxide and lead fluoride.

Recently, the fluxed-melt technique has been successfully applied to the production of ruby and corundum crystals (figure 11). These grow as faceted tabular crystals of reasonably large size and good crystalline shape; they have the added advantage of being strain-free.

OTHER GROWTH TECHNIQUES

In general, most gem materials have such high melting-points and low vapour-pressures that they cannot be grown from the pure liquid or vapour. Two gem materials have, however, been thus grown. Firstly, large fluorite (calcium fluoride) crystals have been obtained by cooling the pure melt, using a method originally devised by P. Bridgman and further developed by D. C. Stockbarger [11]. The essential feature of the process is the limitation of nucleation to a point at the bottom of a specially shaped crucible as the crucible is slowly cooled.

Blende (zinc sulphide) and several structurally related sulphides and selenides have been grown from the vapour phase as hexagonal prisms and plates up to a few centimetres in length. Growth is achieved in two ways: by distillation in vacuo, inert gas, or hydrogen sulphide, and by reaction of zinc and sulphur vapours in a carrier gas. Both methods are reasonably successful, although control of nucleation is not easy.

SPECIFIC PROBLEMS OF CRYSTAL GROWING

To obtain reasonably large crystals from which specimens can be cut for scientific study or for use as gemstones, nucleation must be limited to as few sites as possible. This can sometimes be achieved by the introduction of one or more seed crystals, or by adopting special procedures to initiate growth at one point only. In some techniques neither method is practicable, particularly if high levels of supersaturation or supercooling are developed.

When growth has started, the rate must be controlled so as to obtain good-quality crystals and prevent the formation of other nuclei. In general, the temperature at which a crystal is grown limits to some extent the rates of growth that will give good crystals. Thus, corundum crystals grown by flame fusion can reach a weight of 100 g in three to four hours; from aqueous solutions, at lower temperatures, the time of growth will be months rather than hours; and growth from solution in molten salts at intermediate temperatures requires days or weeks for a product of comparable size.

A second important consideration in crystal growing is purity. The physical properties, particularly optical and microwave absorption, of many materials are modified by quite small amounts of impurities. It is therefore essential to control rigorously the concentration of any desired impurities.

Natural crystals usually contain high concentrations of impurities, and these often cause them to be structurally different from the synthetic products. At a lower level, most 'pure' chemicals used for crystal growing also contain small amounts of such common elements as iron, which may be incorporated in the crystals. It is therefore essential to determine which impurities are present and to take steps to eliminate them, for example by initial purification using conventional methods such as zone refining, fractional crystallization,

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chromatography, or distillation [12]. Controlled additions of impurities may then be made and the properties of the material assessed without confusion. As a consequence, however, subtle distinctions of shade and colour that are found in natural gems may be absent in the purer synthetic counterparts, since the primary colouring is often modified by secondary impurities. The 'pigeon's blood' hue of good natural ruby is due mainly to the presence of chromium, but also to small amounts of iron and possibly titanium. Slight differences in colour produced in this way are, however, usually apparent only to the expert gemmologist.

In most high-temperature processes, impurities may come from the crucible or other container as well as from the materials used. This source of impurity is not important in the Verneuil technique, where the crystal is its own support, but may be very serious when crystals are grown from the melt or from solution. In many cases only platinum or graphite is acceptable as a material for crucibles.

Large concentrations of 'doping' agents—single, controlled, additions—may seriously interfere with the growth processes or may cause modification of crystal habit. In flame fusion, excessive amounts of iron cause severe cracking in the crystals, and in addition make them difficult to grow because of modification of the surface tension of the melt.

INDIVIDUAL SYNTHETIC GEMSTONES

Diamond

Early experimentalists in the field of diamond synthesis attempted to use high-pressure techniques. For example, Hannay heated his hydrocarbon/lithium mixtures in welded iron tubes at red heat. Moissan adopted a different approach, in which carbon precipitated from a molten iron solution was subjected to pressure by the sudden quenching of the mixture. His technique was subsequently repeated by several other workers, but it is now certain that the conditions attained in any of the early experiments could not possibly have led to the formation of diamond.

Although the 'belt' apparatus enabled Bovenkirk and others to reach the diamond-stable region, it was found that the direct conversion of carbon to diamond was too slow. Alternative techniques attempted, including growth from solutions of carbon in metals and by pyrolysis of carbonaceous materials, were not successful. The answer to the problem was found in the use of a metal catalyst to speed the direct conversion. Several metals were found to be suitable (for example nickel, tantalum), and the discoveries enabled appreciable crystal growth to be obtainable for durations of only a few minutes.

The first synthesis of diamond substantiated by X-ray examination was achieved by the General Electric Company of America in 1955. Since then many other laboratories have claimed success, although only a few details have been published [13].

The first specimens appeared as small irregular-shaped black particles, similar in appearance to powdered coke. The progress made in this field will be apparent by the shape of the recent crystals, shown in figure 5. Although crystals of over 0·2 g have been obtained, they are reported not to be of gem quality. Presumably they contain flaws or imperfections; they may well contain the internal dendritic skeleton that is typical of crystals grown rapidly from solution.

The colour of the crystals can be modified by the conditions of growth used, but so far there is no suggestion that 'blue-white' stones have been produced. Nevertheless, it seems certain that with subsequent improvements of both techniques and apparatus, even larger crystals of better quality will be produced. The full effect on diamond prices of the availability of these crystals will certainly be felt when this stage is reached. Initially, the cost of production was rather higher than the market value of natural stones of similar size, and although the synthetic stones can be used for cutting purposes, they may be less strong than natural ones.

Corundum

Corundum is found extensively in nature as small crystals and occurs in many varieties, the best known being the coloured forms, ruby and sapphire. Corundum was successfully synthesized by Verneuil in 1904, and artificial crystals have been grown by a virtually unchanged technique to the present day. They are produced either as elongated cylinders (boules), or as thin rods. The crystals are coloured—red by chromium, yellow by nickel, blue by titanium, green-blue by vanadium; various combinations of these additives give an extensive variety of shades [14]. The uncoloured and pink forms are largely used for jewel bearings for instruments and watches, and for gramophone styli.

The flame-fusion crystals frequently contain small gas-bubbles and sometimes unfused powder. These, together with the colour striations often observed, serve to differentiate the synthetic from

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the natural material. Ruby grown by other techniques may be more difficult to identify by appearance alone, but refined optical-absorption techniques often show small differences between natural and synthetic crystals.

A range of synthetic corundum crystals and cut stones is shown in figures 9 and 12. The flame-fusion crystals are normally unfaceted, although there is some tendency for flattening of the sides to occur if the hexagonal axis lies perpendicular or parallel to the direction of growth.

One of the most interesting developments in sapphire production has been the synthesis of 'star' stones. Crystalline inclusions are frequently found in natural corundum, and, if they are small enough, they may lie along directions related to the crystal axes. This effect is commonly known as 'silk' or 'rutilation', as the included material is often rutile (TiO_2) needles. Under appropriate lighting conditions, diffraction effects are observed as a six-pointed star. If a small amount (0·3 per cent) of titanium dioxide is added to the powder used for growth, it becomes incorporated in the lattice as Ti^{3+}. On heating the crystals in oxygen at moderate temperatures, oxidation to Ti^{4+} occurs with crystallization of rutile needles along three directions perpendicular to the hexagonal axis. At higher temperatures, the Ti^{4+} tends to migrate to the surface of the crystal, and the effect is lost. This crystallization is restricted to layers near the surface of the corundum crystal, and consequently it is necessary to heat-treat the cut stone. Synthetic star rubies and star sapphires are shown in figure 6. These stones are characterized by the perfection of the rays of the star, as compared with natural stones in which the rutilation may be patchy or imperfectly developed.

Spinel

The addition of magnesium oxide to the alumina used for flame-fusion growth causes a change to the spinel structure. Ideally, the crystal would have the formula $MgO.Al_2O_3$, but in practice it is difficult to grow crystals with this composition by flame fusion. The spinel structure persists up to relatively high concentrations of alumina, but a composition near to $MgO.2{\cdot}5Al_2O_3$ is preferred for growth. The spinel thus formed has a defect crystal structure with numerous vacant sites. The incorporation of impurities in such a lattice is often easy and, in fact, spinels can be obtained in a greater colour range than can corundum. Of particular beauty is the cobalt-doped spinel, which has an intense blue colour (figures 9 and 12).

Rutile and strontium titanate

These gemstones have no natural counterparts, as mineral rutile is usually very dark or black and is of limited gemmological value, and strontium titanate does not occur naturally. Both these materials have extremely high refractive indices, with an optical dispersion higher than that of diamond. They surpass diamond with respect to the 'fire' displayed by a cut stone, but are relatively very soft. They are therefore relegated to use as 'costume' jewellery, and, in spite of their attractive appearance, are not popular.

Rutile is difficult to prepare in a perfectly colourless state, and the cut stones frequently show a yellow tint. Furthermore, the crystal structure is tetragonal, and double refraction is readily observed. Both effects serve to distinguish rutile from strontium titanate, which is optically isotropic and is closer to diamond in appearance (figure 7). Coloured stones of both materials have been prepared: specimens of rutile are shown in figure 16. The colour range at present is limited to shades of orange, blue, and green.

Quartz

The hydrothermal production of quartz must represent the most successful synthesis of any gem material. The crystals obtained show a high degree of crystalline perfection and are relatively pure. Production on an industrial scale has commenced in the United States, to provide material for piezoelectric wave-filters.

The crystals are normally grown on seed plates cut perpendicular to the hexagonal axis, as this orientation corresponds to the direction of fastest growth. Growth is normally stopped before a complete bipyramid is formed, as it is desirable to eliminate excessive cutting or wastage of material. It is also preferable to grow a large number of small crystals of uniform size, rather than a few large crystals that would require out-cutting facilities. The process is reliable and extremely reproducible; as it is difficult to observe or measure conditions inside the autoclaves, the control achieved is surprisingly good.

Many coloured natural forms of quartz occur and are used as gem materials. These include amethyst, smoky quartz, rose quartz, citrine, cairngorm, and many other varieties. It is difficult to colour synthetic quartz, and the first green crystals were produced accidentally. The use of potassium compounds, instead of sodium, for the solvent results in the incorporation of iron and other elements from the autoclave wall, giving

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FIGURE 5 – *Synthetic diamonds (by courtesy of the General Electric Company of America).*

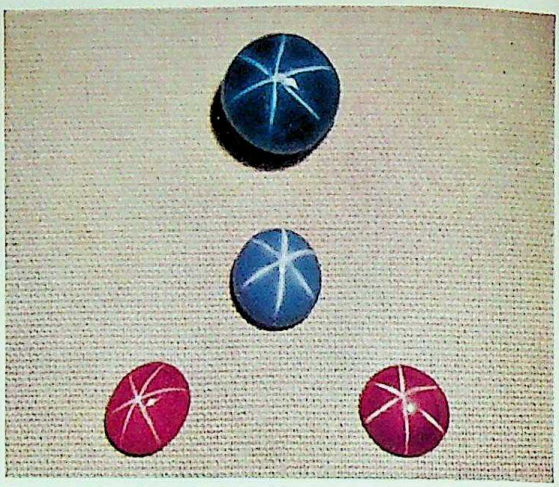

FIGURE 6 – *Star sapphires and rubies.* ($\times 1\frac{1}{2}$)

FIGURE 7 – *Strontium titanate, natural diamond, and rutile brilliants (from left to right).* ($\times 1$)

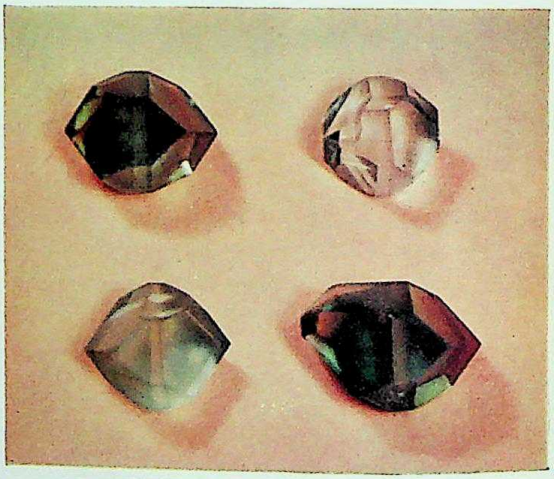

FIGURE 8 – *Coloured quartz crystals.* ($\times \frac{2}{5}$)

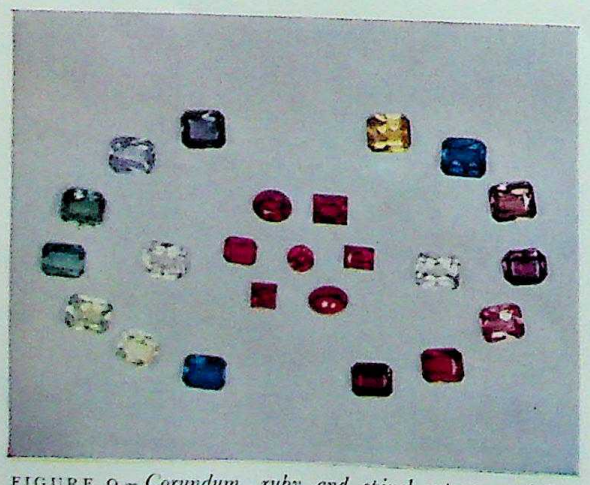

FIGURE 9 – *Corundum, ruby and spinel cut stones (from left to right).* ($\times \frac{2}{5}$)

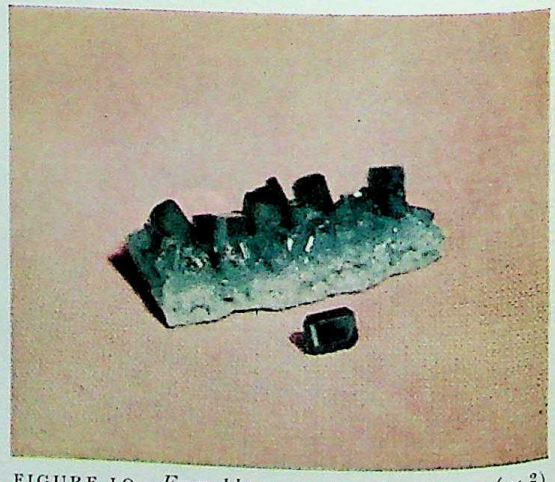

FIGURE 10 – *Emerald; aggregate and cut stone.* ($\times \frac{2}{5}$)

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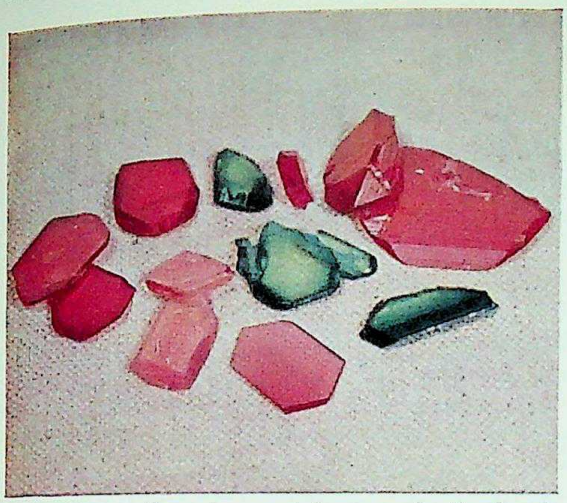

FIGURE 11 – *Corundum crystals grown by the fluxed-melt process.* ($\times \frac{2}{3}$)

FIGURE 12 – *Corundum (top row) and spinel boules.* ($\times \frac{1}{4}$)

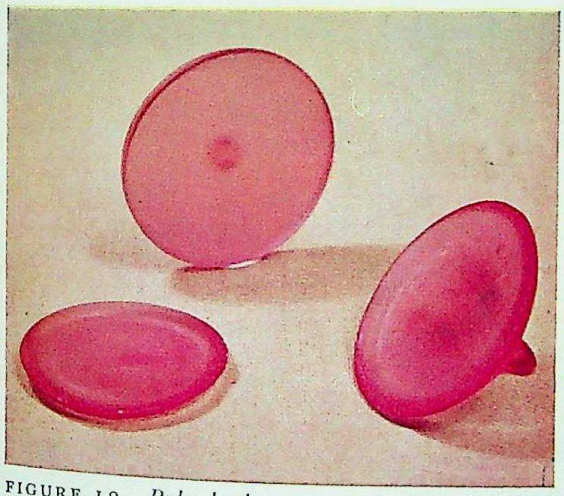

FIGURE 13 – *Ruby boule grown by 'spinning' (sliced across).* ($\times \frac{2}{3}$)

FIGURE 14 – *Quartz crystals.*

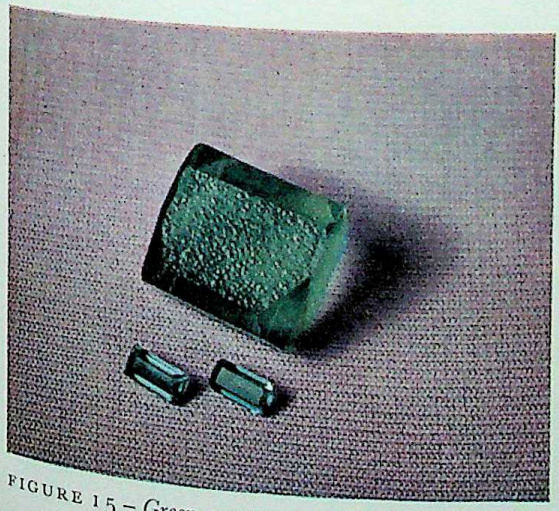

FIGURE 15 – *Green quartz crystal and cut stone.* ($\times \frac{2}{3}$)

FIGURE 16 – *Rutile cut stones.* ($\times \frac{1}{2}$)

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crystals coloured green or brown. It is likely that other colours could be produced, but it would be necessary to grow the crystals in inert liners to prevent contamination with iron. Crystals of both colourless and coloured quartz are shown in figures 8, 14, and 15.

It is perhaps surprising that amethyst has not yet been synthesized, but so far no distinct red or mauve colouration in quartz has been described, apart from that produced by irradiation. It is thought that the colour centres in amethyst are associated with disordered structures produced by irradiation from nearby radioactive materials.

Emerald

The history of emerald synthesis goes back many years, but progress has been relatively slow. Although small crystals of beryl, $BeO.Al_2O_3.6SiO_2$, were produced in 1888, emerald crystals (chromium-bearing beryl) of appreciable size were not forthcoming until the appearance of the German 'Igmeralds' in 1934. The most successful synthesis, that is, the one giving the highest-quality stones, was by Caryl Chatham in the United States. He did not divulge his process, and these emeralds have consequently remained an enigma for over 20 years. The growth process is reported to be a lengthy one and is generally considered to be hydrothermal, although the success of the fluxed-melt technique for other materials makes this also a method to be considered. The crystals can evidently be grown on natural seeds, but examination of 'rough' clusters of crystals suggest that they can also develop from small spontaneously nucleated crystal-clusters. Only the Chatham process can be considered to be industrially successful, since 'Igmeralds' were not of good crystal quality, and development of the process was stopped by World War II.

Garnet

Natural garnets are currently not popular as gemstones, although a number of mounted stones prized as rubies are in fact probably garnets. The appeal of the garnet family was undoubtedly in the wide range of colours obtainable, which includes good greens and reds, and black. Both the structure and chemical composition of natural garnets are complex. The basic formula, $A_3B_2C_3O_{12}$, allows a wide variation of valency and ionic size for each of the three available lattice sites, which differ from one another in the co-ordination with the surrounding oxygen ions. In most natural crystals the 'C' sites are predominantly occupied by Si^{4+}, with 'A' sites occupied typically by Ca^{2+} or Fe^{2+}, and 'B' by Fe^{3+} or Al^{3+}.

The synthetic garnets are in general of simpler composition. The first compound to be grown as large crystals (1 cm diameter) was $Y_3Fe_5O_{12}$, in which iron occupies two of the available sites. This arrangement leads to interesting magnetic properties that are due to the interaction of the ions in the different sites, and considerable attention has been given to this compound and to the related crystals containing rare-earth and other paramagnetic ions.

The yttrium-iron garnets are black lustrous crystals similar in appearance to the natural garnet melanite. The crystals are transparent when viewed in thin sections, and the magnetic domains (regions in which the magnetic effects cooperate in a single direction) can be viewed directly under the polarizing microscope.

Compositions of the type $Ga_3Al_5O_{12}$ and $Y_3Al_5O_{12}$ have also been grown by fluxed-melt techniques as transparent polyhedral crystals. These can be coloured to give pleasing effects by the addition of various transition-metal oxides.

Garnets closer in composition to the natural species have been produced under ultra-high pressure; for example, the natural form andradite, $Ca_3Fe_2Si_3O_{12}$, has been synthesized as small crystals.

Blende

Crystals of blende have been successfully grown by the vapour-phase technique. The crystals are obtained as hexagonal prisms, or occasionally as plates or needles—the form obtained depending on the conditions of growth. Natural blende crystals are usually dark, and the substance is of limited interest for gemstones because it is soft, although the optical dispersion is high. The structural transition from cubic to the hexagonal crystal system occurs at a relatively low temperature and the hexagonal form is usually obtained, although both types of structure may be present in disordered specimens.

Minor gems

Small crystals of many of the minor gems have been grown, particularly by hydrothermal means. There has been little attempt to produce larger crystals, although this would certainly be possible in many instances. The present emphasis on single crystals for investigations in solid-state physics may lead to further development of methods for growing these materials, but there is little incentive to

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produce crystals for gem use, as the minor gemstones have no outstanding characteristics.

SCIENTIFIC USES OF GEMSTONES

It would be convenient to be able to predict in detail the full physical properties of a compound from a knowledge of the crystal structure alone, but at present too many unknown factors are involved. Certain predictions can be made by analogy with similar structures, but we are only just beginning to understand the physical processes involved in energy transfer in crystals. This situation may partly be due to the fact that perfect crystals cannot be grown. Apart from gross imperfections such as cracks, inclusions, or misorientation, the importance of dislocations on the physical and mechanical properties has become increasingly apparent in recent years. In fact, as measurement techniques become more refined, the need for more nearly perfect crystals becomes acute.

Applications of synthetic gemstones

Application	Purpose	Crystal	Trace additives of interest
Paramagnetic studies	Fundamental research	Corundum Rutile Emerald Garnet Fluorite Spinel	Transition metals, rare-earth and actinide elements
Masers	Low-noise amplifiers; microwave generation	Corundum Spinel Rutile	Fe, Cr Fe, Cr Fe, Cr
Lasers	Coherent light or infra-red high-intensity source	Corundum Fluorite	Cr U, Sm
Fluorescence ..	Light sources; scintillation counters	Corundum Fluorite Blende Zincite	Cr Mn Cu, Ag, Mn
Photoconductivity, photoelectricity, semiconduction	Fundamental studies	Diamond Blende Zincite	Cu, Ag
Ferrimagnetic studies	Microwave devices; fundamental research	Garnet	Various magnetic and non-magnetic substituents
Optical materials	Lenses; high-dispersion prisms; U.V. or I.R. transmission windows	Strontium titanate Corundum Fluorite Quartz	Pure
Piezoelectric devices	Wave filters	Quartz	Pure
Miscellaneous uses	Refractories; insulators; dielectrics; abrasives	Corundum Spinel Rutile Garnet	

Much of the research with crystals has been undertaken with materials that are currently easily available, and these are not necessarily the most suitable ones for study. As the shortcomings of such materials are discovered, demands are made upon the crystal grower for new types or modifications of crystals.

The existence of processes for the synthesis of gemstones has made these materials an obvious choice for the study of crystals (see table), particularly in terms of electron-spin resonance. The important properties in this connection are the discrete energy-levels possessed by paramagnetic ions distributed evenly in a stable host-lattice. The energy exchanged in transitions between the ground and excited states may correspond to electromagnetic radiation frequencies ranging from the centimetric region through to visible light, depending on the type of ion present, the effect of the crystal environment, external magnetic fields, and factors related to the perfection of the specimens.

Although water-soluble crystals have been used for these studies, more durable crystals are preferable. Since the colour of gem materials is often produced by paramagnetic impurities in concentrations of less than one per cent, gems are particularly suitable for this work. The natural stones usually contain several impurities that complicate the interpretation of the results, and synthetic stones with a single controlled additive are consequently more useful for study.

A device making use of energy transitions in the electron-spin

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resonance spectrum is the maser, which has been described in detail elsewhere [15], and is also discussed in this issue of ENDEAVOUR.

Both masers and lasers have been made with ruby as the working substance. However, the imperfections of the crystals grown by flame fusion detract from their performance, and crystals grown by slower or more controllable processes are potentially more useful. Many other types of crystal have been investigated for these applications (see table), including emerald, spinel, and rutile. Of these, spinel and rutile are particularly suitable for maser operation at higher frequencies because of favourably disposed energy-levels associated with the particular crystal structures.

A second field of study, which also involves energy transitions in the visible region, is that of luminescence. This general term includes a number of processes in which the emission of light is produced by the application of electrical or mechanical energy, or electromagnetic radiation. Unlike the laser, the emission in this case is spontaneous and not stimulated. Again, impurity ions in the crystal lattice are important, as they provide a means of modifying the conductivity-band structure by the introduction of extra energy-levels or electron-traps. The nature of the modification depends on the valency, electronic structure, and concentration of the element, and on the way it is incorporated into the crystal lattice. The energy exchange associated with various types of electron transition determines the nature of the phenomenon shown. In all types of luminescence, the release of energy is *ipso facto* at frequencies near to that of light, and the various processes differ in the way in which energy is absorbed. These may be, for example, electron bombardment by the application of an electric field, ultra violet irradiation, or mechanical damage. These processes are closely allied to the phenomena of photoconductivity, photoelectricity, and semiconduction. Blende has proved to be a particularly useful crystal for the study of many of these effects, and it is also the only substance yet found which produces a useful amount of light by electroluminescence. 'Cold light' panels using zinc sulphide are manufactured for applications such as display notices, that require prolonged life with only a low light-output.

There is often a considerable overlap between the various aspects of fundamental research, and information gained in one field may be supplemented by studies in entirely different ones. Gradually an overall picture relating energy processes to structure and structural imperfections is emerging, and the results of these labours should be reflected in improved materials and devices for domestic use as well as in industry and research. In the meantime, the creative abilities of the crystal grower are still much in demand, and synthetic gems are likely to continue to play a dual role for some time to come.

ACKNOWLEDGMENTS

The author wishes to express his thanks to Dr G. F. Claringbull of the Natural History Museum, London, for permission to photograph the specimens of synthetic gemstones shown in figures 6, 7, 10, 12, and 16; and also to Mr H. Ginder of Messrs J. and C. Ginder, Hatton Garden, London, for the loan of the fine-quality diamond shown in figure 7.

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Morphogenesis in plant tissue cultures

J. REINERT

Tissue cultures of higher plants differ from those of higher animals in that they retain their ability to generate new organs, both roots and, more rarely, shoots. These organs will develop even in tissues that contain no rudimentary organs. This article describes the conditions under which the new organs develop, and some experiments on the factors that determine whether it is roots or shoots that actually do develop, and on the effects of chemicals in deciding generally the particular type of new cell that forms.

Ever since it became possible to grow tissues of the higher plants *in vitro* they have been used to study the origin of form. Plant cells and tissues are excellent materials for the purpose because—unlike those of higher animals—they often retain their ability to form new organs (leaves, stems, and roots) even in culture.

Although the significance of cell and tissue cultures for plant physiology had already been recognised and pointed out by G. Haberlandt [1] at the turn of the century, it was not until 1938–39 that R. J. Gautheret [2], P. Nobécourt [3], and P. R. White [4] independently succeeded in culturing normal and tumerous tissues from various plants over indefinite periods, and so opening up a new field for physiological experiments of the most varied kinds. This success was due mainly to the replacement of the mature and largely differentiated cells, with which Haberlandt and his associates had worked, by tissues that contained embryonic cells. It was also important that synthetic auxin (3-indoleacetic acid) was already available at that time.

In these first successful experiments, relatively simple, chemically unambiguous media were employed: they contained nutrient salts, trace elements, sucrose, and also a few vitamins, individual amino acids, and auxin. Tissues from the most various plants and organs could be cultured on these simple media (figure 4). By far the greatest proportion of the cultures, however, originated from the dicotyledons. With other groups there were considerable difficulties, as they demanded more elaborate culture media. Up till then, they could be grown adequately only if the simple synthetic media were supplemented with more complex components of unknown composition. The most effective and most frequently used of these was coconut milk; but yeast and malt extracts also proved suitable. Tissues from the gymnosperms gave results similar to those from monocotyledons. The difficulties with cultures from various coniferous species are very well known; few of them grow on synthetic culture media and the majority can still be sustained only on media that contain coconut milk or malt extract.

The formation of organs has been observed in tissue cultures from all the groups of plants mentioned above. Root formation is the most frequent (figures 2, 5, and 10), both on tissues from roots and on those from shoots. The roots are distributed irregularly over the culture and may be entirely normal in their structure. The expression normal here applies, however, only to the root meristem and the growth and differentiating zones lying behind it; the base ends blindly in the parenchyma of the tissue culture [5] (figure 2). When isolated and grown in suitable nutrient solutions, these roots scarcely differ from those of intact plants.

As a rule, the formation of roots does not depend on the presence of pre-existing rudiments. The fact that the rapidly growing root initials cannot be demonstrated for at least one or two weeks after the isolation of the tissue points to new formation. Initiation and development are two separate phases of growth. During the first, which begins shortly after the isolation of the tissue, there is an acceleration of cell division and a consequent formation of a mass of undifferentiated cells, callus. During this phase they may arise from meristematic, but also from mature, highly differentiated cells, deriving, for example, from the

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phloem. In the second phase there are renewed local accelerations of growth, due to further cell-division, and small groups of embryonal cells are laid down that rapidly organize themselves into growing points (figure 1). The polarity of these root initials, which most often occur in the outer cell layers of the cultures, is indicated at an early stage by the direction of the mitotic spindles.

The above pattern of growth has been consistently established in various tissue cultures of short duration [5]. There are only occasional, less exact observations available on the same processes in callus tissue that has already undergone numerous sub-culturings. The formation of small embryonic centres, foreshadowing root formation, is, however, also evident in these cultures, which often contain fully differentiated phloem and xylem cells as well as the mass of parenchyma.

Leafy shoots develop on tissue cultures much more rarely than roots. Their morphological and anatomical characteristics have been investigated thoroughly in only a few cases. It has, however, been established that rootless shoots of normal structure may be formed in tobacco [7] and *Sequoia* [8] tissues, for example. Cultures from carrots may develop complete plantlets corresponding closely in their young stages with those raised from seed [6]. In one case it was even possible to separate these carrot plants from the culture and to bring them on to the stage of fruiting [9]. On the other hand, structures arising on tissues derived from *Agave* can be described only as leafy or bud-like [10].

The first stages of development of the shoots and leaves do not differ from those that have been described for roots. In newly isolated cultures, undifferentiated parenchyma, that can have its origin in different types of cells, arises in the first phase. During the second, developmental, phase, which can be observed even in callus cultured for years, the initiation of the embryonic centres occurs in the parenchyma having a relatively high rate of cell division; shoots and leaves develop from these centres [5, 7]. During the growth of the initials, cells from neighbouring zones can also be drawn into the meristem of the developing organ [8].

The development of complete plantlets in carrot cultures (figures 8 and 9) follows a rather different course. In certain strains of these cultures, roots are first formed, and the shoot develops on these later [11]. By contrast, a developmental sequence has been observed in other strains which, with its structures akin to egg cells (figures 11 and 14) with proembryos (figure 15) and sundry other stages of embryo and seedling development (figures 12 and 13), shows a surprising correspondence with the stages which follow the fertilization of egg cells [6]. A similar development has recently been demonstrated in callus originating from dodder (*Cuscuta*) embryos [12].

It is not yet possible to make extensive generalizations about the reactions of tissue cultures in organogenesis. However, it is certain that the inception of organs occurs most frequently in recently isolated tissues, and that the ability decreases with increasing duration of culture, and eventually disappears completely; but even these rules correspond only partially with the actual circumstances. Thus, while many cultures remain indefinitely without any organization, others appear to have an enduring capacity for forming organs. The best known examples of the latter type are cultures from the stinkhorn (*Amorphophallus*) [13], and carrot tissues [14].

These alternations do not by any means exhaust the possibilities of variation. Shoots develop on tissues from certain trees (lime, poplar, birch) only when they have their first subculture behind them [15]. It has also been observed that on elm tissues, only shoots developed at first, but after several transitions hardly anything but roots were formed [15]. Some of the most extreme cases have been reported with pathologically modified tissues. In cultures from virus-induced tumours that had grown entirely without differentiation for years, the initiation of roots occurred suddenly, without any alteration of the experimental conditions [16]. Crown gall tissues from *Scorzonera* behaved in the same way; they remained undifferentiated for a long time and then—wholly unexpectedly—formed shoots and roots [17].

There is as yet no really satisfactory explanation for this behaviour. It becomes less surprising, however, when one considers that plant-tissue cultures are complex systems that can change during their lifetime. This is indicated by many observations, some of them made long ago, on the variable chromosome-numbers of cells in cultures from normal and tumorous tissues (figure 3). It has been made particularly clear by recent cytological investigations. The newer work has demonstrated with high probability that the occurrence of polyploid cells in the cultures depends not only on the division of corresponding cells in the tissue originally isolated but on the conversion of diploid cells. This is very clearly shown in cultures from the diploid meristem of pea roots. It is evident that, under certain

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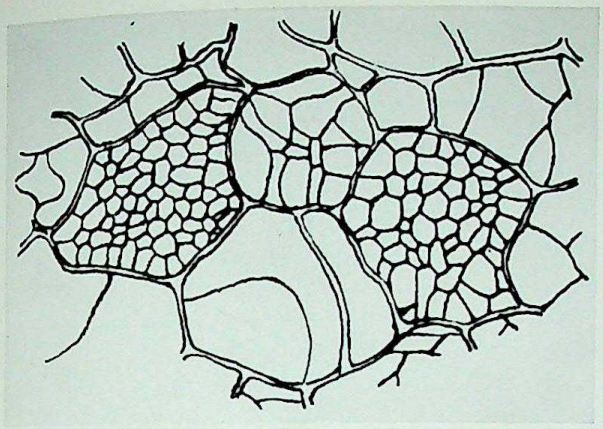


FIGURE 1 – *Cell division in parenchyma cells of a carrot culture. These limited meristematic zones develop later into root meristems.*

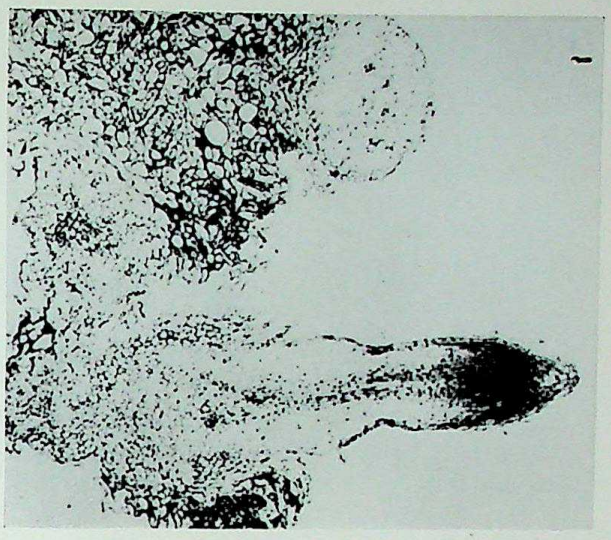
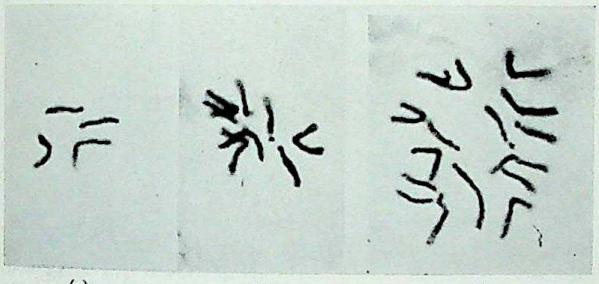


FIGURE 2 – *Median section through a young elongating root on a tissue culture from pea roots.* (×40)



(a) (b) (c)

FIGURE 3 (*left*) – *Metaphases from dividing cells in tissue cultures from* Haplopappus gracilis. (*a*) $2n = 4$; (*b*) $4n = 8$; (*c*) $8n = 16$. (× 1330)

experimental conditions, only the diploid cells of the meristem are dividing; but only a few days after isolation, among many diploids there are also a few tetraploid nuclei. In the course of a few weeks the rates shift drastically in favour of the tetraploids, and then octoploids turn up and occasional cells with an even higher ploidy [18]. The instability of these cells is not an isolated phenomenon, for similar occurrences have also been observed during the culture of haploid pollen grains [19] and triploid endosperm [20].

A point that must now be more fully discussed concerns the nutrient media. They all contained either yeast extract or coconut milk, ingredients that favour the formation of polyploid cells. Comparison of cultures of *Haplopappus gracilis* on synthetic media with those on media with coconut milk added (figure 3), show that the effect of the complex components must be acceleration of a process that is also initiated by the synthetic media themselves after some time [21].

These considerations limit investigations into one of the most interesting sectors of the work, the control of morphogenesis by manipulation of external factors. Because of the instability of the cultures, all such investigations have employed either recently isolated cultures, or the few whose capacity for forming organs is retained over a longer time.

The effect of physical factors on organogenesis has been investigated really thoroughly on only one object, the tissues from genetically determined tumours on the hybrids of certain tobacco species (*Nicotiana glauca* × *Nicotiana langsdorffi*). Explants from these tumours grow on simple synthetic media as undifferentiated callus. As soon, however, as they are submerged in liquid media of the same composition, they form abundant buds and leafy shoots [22]. Weak light, and temperatures below 25° C, reinforce the effect of the fluid medium; higher light-intensities and temperatures have the reverse effect [23].

It can be assumed that these varied reactions of the tissue depend on the modification of a complex system of various interacting factors. Moreover the same factors may act co-operatively under certain experimental conditions and antagonistically under others. Experiments that have been made with tissue cultures from the stems of tobacco plants are the most conspicuously successful illustration of this [24].

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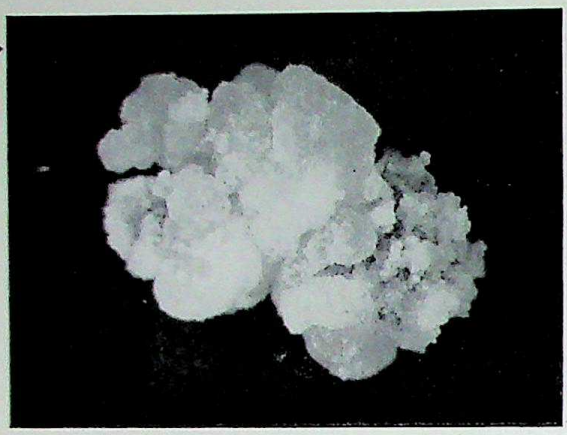

FIGURE 4 – *Tissue culture from carrots.* (×2)

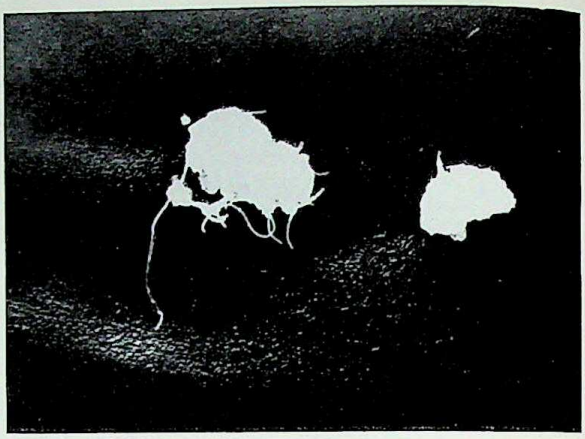

FIGURE 5 – *Root formation on carrot cultures.* (×1)

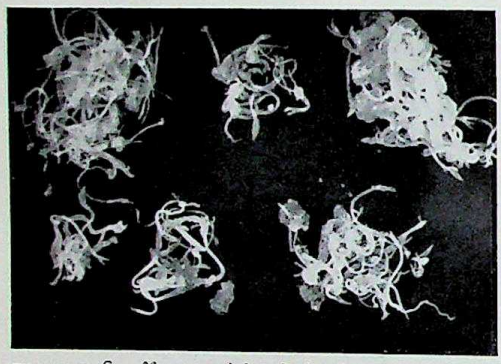

FIGURE 6 – *Young, etiolated plantlets on carrot cultures.* (×½)

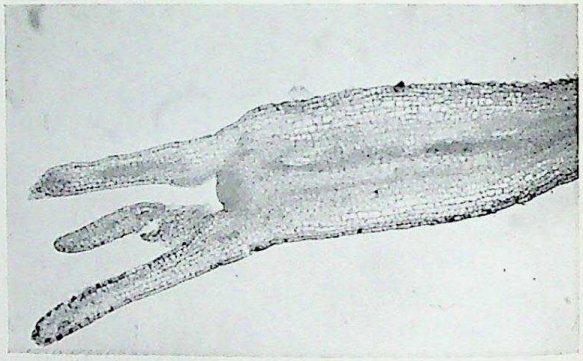

FIGURE 7 – *Median longitudinal section through an adventitious embryo from the marginal zone of a carrot culture. The outermost tip of the root cap broke off during withdrawal from the culture.* (× 55)

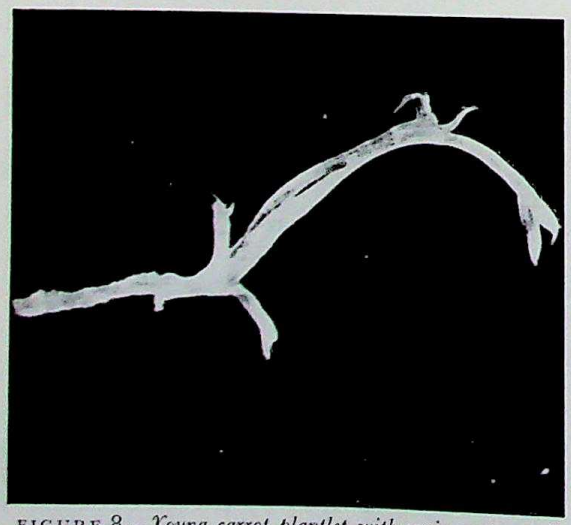

FIGURE 8 – *Young carrot plantlet with main roots, cotyledons and first leaves removed from a carrot culture.* (×2½)

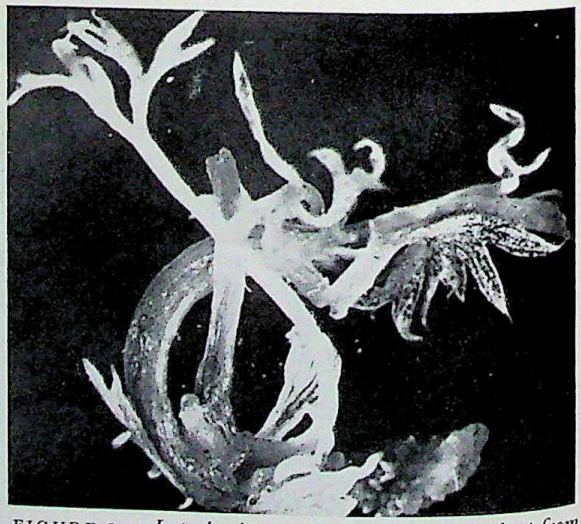

FIGURE 9 – *Late developmental stage of a carrot plant from a culture which was illuminated.* (×2½)

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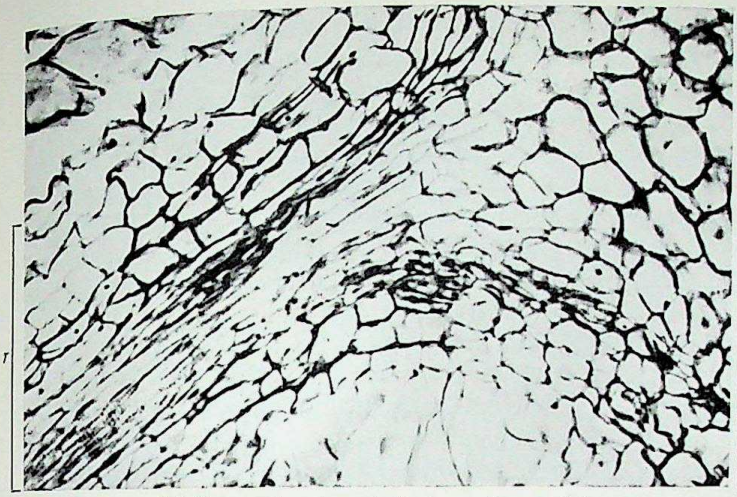


FIGURE 10 – *Median section through the base of a root (r) on a carrot culture. In the older zones of the root tracheidal cells of the conducting strand are recognizable.* (× 200)

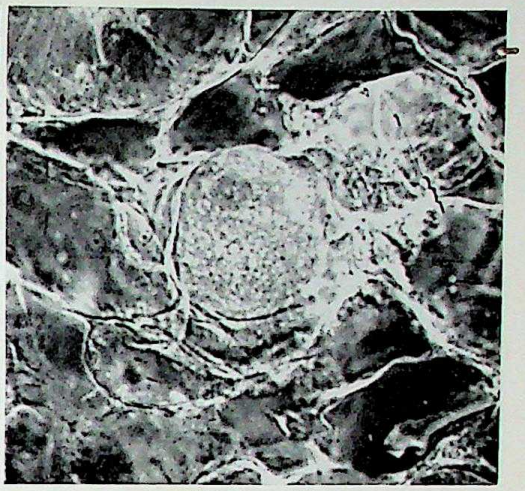


FIGURE 11

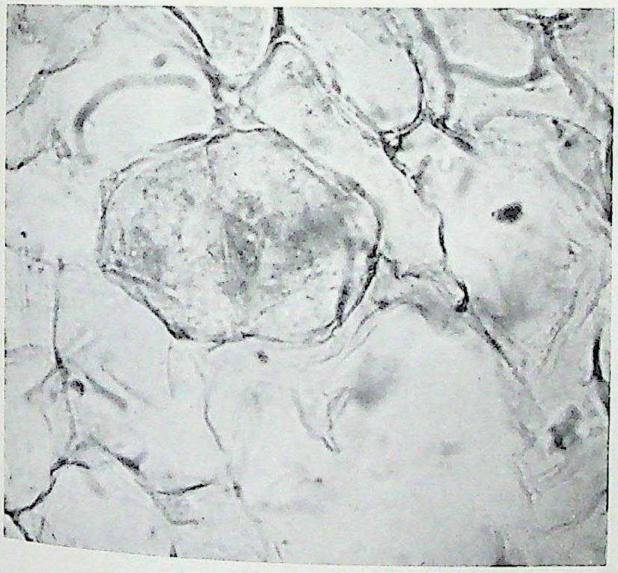
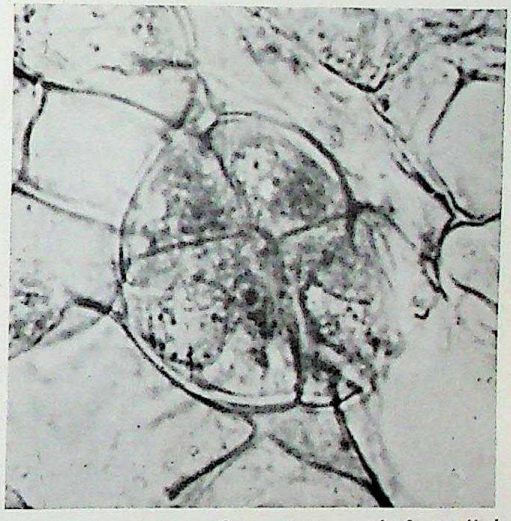


FIGURE 12



FIGURES 11–13 – *One-, two-, and four-celled stages of embryonic cells, formed in parenchyma of carrot cultures.* (× 760)

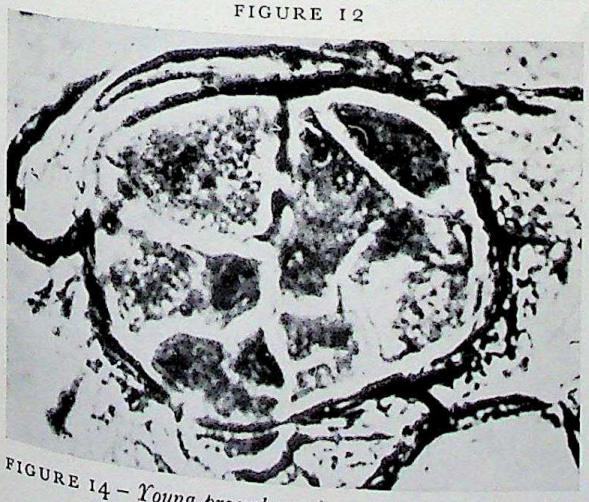


FIGURE 14 – *Young proembryos in carrot cultures.* (× 180)

FIGURE 15 – *Structures resembling egg cells in the parenchyma of carrot cultures from which adventitious embryos develop later.* (× 760)

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When these cultures originate from the pith parenchyma alone, cell division and true growth occur on a synthetic medium only if it contains auxin and kinetin (6-furfurylaminopurine). These two substances determine, however, not only whether the tissue grows, but also how it grows. If, for example, the nutrient medium contains auxin and kinetin in concentrations of 2 mg per litre and 0·1 mg per litre respectively, only an undifferentiated callus results. The two compounds in this case work additively, as is shown by a high growth rate. If, however, the kinetin concentration is lowered to 0·02 mg per litre without altering the auxin level, roots arise abundantly in the cultures. Higher concentrations of the purine derivative (0·5 mg per litre) lead, conversely, to the initiation of shoots, and root formation is then entirely suppressed.

Other compounds can interfere in this antagonism. Raising the phosphate concentration of the standard medium five or six times can—like the additions of kinetin—weaken or suppress the root-promoting effect of the auxin and reinforce the shoot formation. Adenine, casein hydrolysates, and individual amino acids, such as tyrosine, act similarly. In equivalent concentrations, they augment the kinetin effect and reduce that of auxin.

The same mechanism, in which shifts of the ratio of the concentrations of auxin and purine derivative lead either to root or shoot formation, has also been proved for cultures from other plants. Other tissues behave differently. Of these only carrots and pea tissues will be mentioned here. In these, auxin—in contrast to the observations on tobacco tissues—suppresses all organ-formation, which can then be achieved only by transfer to auxin-free media [6, 17]. There are not yet enough experimental results for the significance of this behaviour to be clear; there is nevertheless enough evidence from the investigations with the carrot cultures just mentioned to show that growth and organ-formation are regulated by a complex system. In this, besides auxin, amino acids and other factors play a part.

In judging the value of what has been discovered about morphogenesis it must be remembered that the subject is still young. Certain aspects of it can now be examined under strictly controlled conditions and with chemically defined media. The idea that the formation of organs depended on a specific organ-forming substance called caline was an old dogma highly esteemed until recently. It was overthrown, and could be overthrown, only by work with tissue cultures. Intact plants do not provide such good materials for this sort of experiment.

The most immediate difficulty to be overcome in the use of tissue cultures for physiological investigations arises from the variability of the cultures. Only when their characteristics can be held stable will they become the ideal materials for such experiments.

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The early development of analytical chemistry

F. GREENAWAY

The history of analytical chemistry has not been much studied until recently, although simple analysis, in the form of assaying—the testing of the purity of silver and gold, has been practised for some thousands of years. This article discusses some aspects of the development of analytical chemistry and its connexion with the development of chemical theory in the period that culminated with the great advances of the eighteenth and early nineteenth centuries.

Analytical chemistry is normally rather neglected in histories of science, partly because its progress lacks the excitement of the changes associated with, say, Lavoisier and Dalton, and partly because it seems, at first sight, that progress in analysis must be dependent on progress in chemical theory. This neglect is unjustified: analytical chemistry played a part in the progress of chemistry that is neither secondary nor derivative. Some of the most important features of chemical thought first appeared as a result of analytical practice, as will be shown, and many ideas found confirmation in their application to new systems of analysis.

It is not possible in so short an article as this to consider more than one line of development; a suitable one ended with Gay-Lussac's monograph, published in 1832 [1], on the determination of silver by titration. The monograph shows traces both of the traditions of assaying and of the innovations of theory and analytical practice of the preceding century.

ASSAYING

Tests of the quality of goods are as old as trade itself, and many have been recorded. Pliny records tests of such substances as nitre, minium, vitriols, and indigo, by smell, taste, feel, appearance, effects of heat, and density. These are not analytical techniques, but an equally early technique, that of assaying, certainly is. It is both the oldest quantitative chemical technique and one of the most fundamental.

It has been known for a long time that the ancient civilizations used cupellation—the process by which the purity of silver or gold is determined by heating a sample with lead in an absorbent crucible or cupel. The lead and the impurities are converted into a slag, which is absorbed by the cupel and leaves the silver or gold behind. It has been shown [2] that this process was well established in the ancient Middle East. It travelled from there, probably by more than one path, and arrived eventually in western Europe. The technique was considered by the twelfth century to be a traditional art, and was widely used after that date.

An early example of the interest it held for the man of affairs is to be found in the 'Dialogue of the Exchequer' (*Dialogus de Scaccario*) of Robert Fitznigel [3]. This is a treatise on the structure and functions of the Court of Exchequer and dates from about 1180. Robert admits that he is no metallurgist, but he describes the procedure clearly.

'The pence are counted in the presence of the Melter and of the persons sent by the Barons and are handed to the Melter. He takes them, counts them in his own hands, and arranges them in an ash cupel in the furnace. He then, following the rules of his art, melts them into an ingot, fusing and refining the silver . . . when the assay is completed the Knight Silversmith, accompanied by the others, brings the ingot to the Barons, and weighs it. He then makes good what the fire has consumed by adding the pence from the same purse till the test-piece balances the pound. It is then marked with chalk as follows: "Yorkshire".

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The pound lost in the fire *so many* pence; and then it receives the name of "Assay".'

In answer to the question 'What are the indications whether an assay is or is not complete?' he says 'I do not really know because I have never studied the subject. But so long as a black film is seen floating about over the now liquid silver, the assay is said to be unfinished. When, however, small grains appear to rise from the bottom to the top, and there dissolve, it is a sign that the assay is complete'.

This is of interest as technical reporting, of course, but more interesting is the implied context, an administrative system making use of a sophisticated chemical technique.

By the fourteenth century the technique was so much taken for granted that Nicolas Oresme in his treatise *De Moneta* [4] could write as if the techniques were common knowledge among his readers, referring to them in support of his arguments, and assuming an acquaintance with the chemical detail. He also suggests that the community should protect itself against debasement of the coinage (through surreptitious changes in the alloy) by establishing standards with which suspect coins could be compared. This idea of a legal standard was already old in his day, trial plates having been mentioned in England at least as early as 1248 [5]—the oldest known survivor is the Silver Trial Plate, of 1279, in the Royal Mint [6]. These discussions imply a widespread trust in assaying.

Such trust was not blind confidence in a set of arbitrary techniques. A note of about 1300 instructs the assayer to make three tests, lest through overheating, loss of silver, or failure of the fire there should be an error in one of them. If unequal weights are found, the heaviest must be taken as the correct one; because, while silver may be lost, it could not be gained accidentally.

In the fourteenth century, again, we find striking evidence that the assayers understood at least part of the transformation that went on in their cupels. An English Mint document of about 1350 says that to refine silver, 'sterile' lead (lead not containing silver or other metal) should be used. The significance of this is seen in a French edict of 1343, which enjoins assayers to carry out a preliminary assay on a specimen of the lead to be used so as to determine its silver content. This would cause a high result were no allowance made for it.

THE RENAISSANCE METALLURGISTS

Many of these medieval notes and interpolations were written by the employers of the assayers, not by the assayers themselves. The earliest records known to have been written from direct observation date from the early sixteenth century.

The *Probierbüchlein* [7] of about 1510 is the first printed text containing chemical techniques recognizably antecedent to techniques still in use today. It was followed by a number of substantial works; several of the more important of these are available in excellent modern editions. Both Agricola's *De Re Metallica* and Biringuccio's *De la Pirotechnia* [8] are delightful to read, as is a work almost entirely devoted to assaying, Lazarus Ercker's 'Treatise on Ores and Assaying' of 1574 [9]. This is an elaborate technical survey, and its range includes base metals and saltpetre, in addition to the noble metals, gold and silver, that were the main subjects of the earlier manuscripts. The text gives a number of general precepts for the assayer. Thus, though he may buy his apparatus, he should know how to construct it, if need be. The instructions are, in fact, so clear that a replica of a sixteenth-century balance has recently been made from them (figure 1); the steel used was actually a sword-blade, and the balance arm swings only when it is raised by pulling forward the piece of wood seen in the centre of the picture. Ercker also states as principles that sampling must be systematic, that records must be carefully kept, and that the assayer who would make a good living must not neglect the art of book-keeping.

Several of these early books ran through many editions, but, although progress in metallurgy led to their elaboration during the seventeenth century, they remained practical handbooks. A new approach is seen in the work of Johann Andreas Cramer (1710–77), one of the many men of science who flourished at the University of Leyden. His principal work is *Elementa Artis Docimasticae* of 1739. The fact that Cramer worked in a university may explain why the book is the first in which techniques of assaying are linked with the wider field of chemical knowledge. It owes a great deal to the influence of the teaching method of Boerhaave, and follows the form of his *Elementa Chymiae* closely. There are two parts; the theory is set out in the first. In it he introduces an important idea, that of the menstruum. He describes this as the medium in which chemical reactions took place, and which indeed rendered them possible, by separating parts of one substance so that they could enter into new relations with parts of another.

In the second part, 'processes', practical methods, are described, that deal with the assaying

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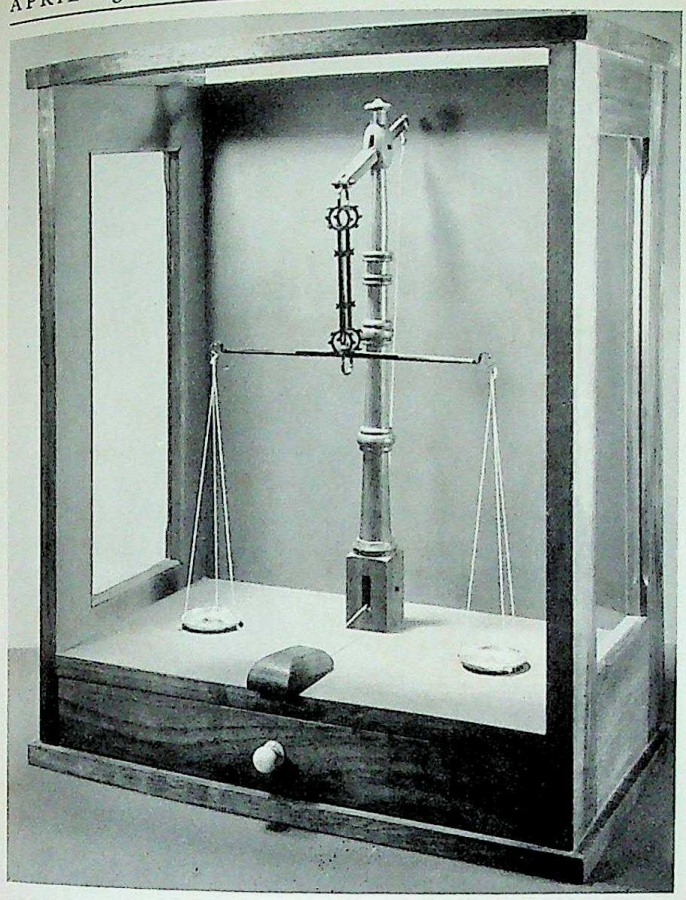

FIGURE 1 – *Reconstruction of sixteenth-century balance for assaying, after Ercker and Agricola. (Science Museum photograph, Crown Copyright reserved.)*

of silver and gold, and with the examination of base metals. It covers a considerable range of mineral chemistry, but there was no theory of the constitution of matter to raise the examination of base metals to the level of that of the assay of precious metals, where, of course, the pure metal remains at the end of the assay. Nevertheless, the book helped to foster a critical study of practical methods that contributed to the developments of theory in later years of the century.

A new approach to the study of the process of cupellation was made in 1760–62 [10]. Two official French assayers had disagreed on the assay of an ingot of silver. Matthieu Tillet was one of the committee set up to investigate the discrepancy. He wrote several papers, independently of the committee's report, in which he described work he had done over a long period. One source of error he had found, by assaying a used cupel, was absorption of silver by the cupel itself. This is a very early instance of the quantitative examination of effects of analytical apparatus on the results of an analysis. Other parts of his work, such as the invention of the ingenious pyrometer with which he was able to establish the consistency and reproducibility of furnace temperatures, and his study of the changes in the lead used, show that Tillet was able to approach assaying not just as a traditional method requiring refinement in technique but as a problem in chemistry requiring the study of each component, process, and physical factor. However, it was too soon for the implications of his work to be appreciated; the phlogiston theory was generally accepted, and there was no accurate idea of the nature of chemical elements and of chemical combination. It was not until after the work of his great French and Swedish contemporaries that the critical study of analytical methods became widespread.

THE BEGINNINGS OF ANALYSIS IN SOLUTION

The metallurgists worked with dry components, but they were not limited to the use of the furnace and the crucible, or even to studies of the extraction of metals. For example, the introduction of the blowpipe made it possible to study small quantities of material quickly and economically. However, the great advance in analytical chemistry came about through the recognition that chemical change could take place through the intervention of a 'menstruum', and particularly through reactions taking place in aqueous solution.

The early assayers had always used other tests than a simple assay. Ercker leached saltpetre-bearing earth and weighed a quantity of liquor, evaporated it to dryness, and weighed again. Libavius describes this method [11] and also the comparison of the weight of a certain volume of

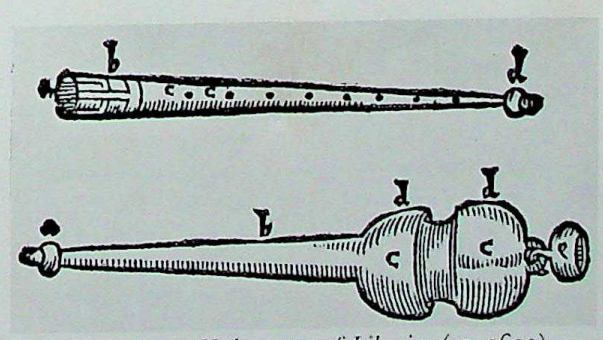

FIGURE 2 – *Hydrometers of Libavius (ca. 1600).*

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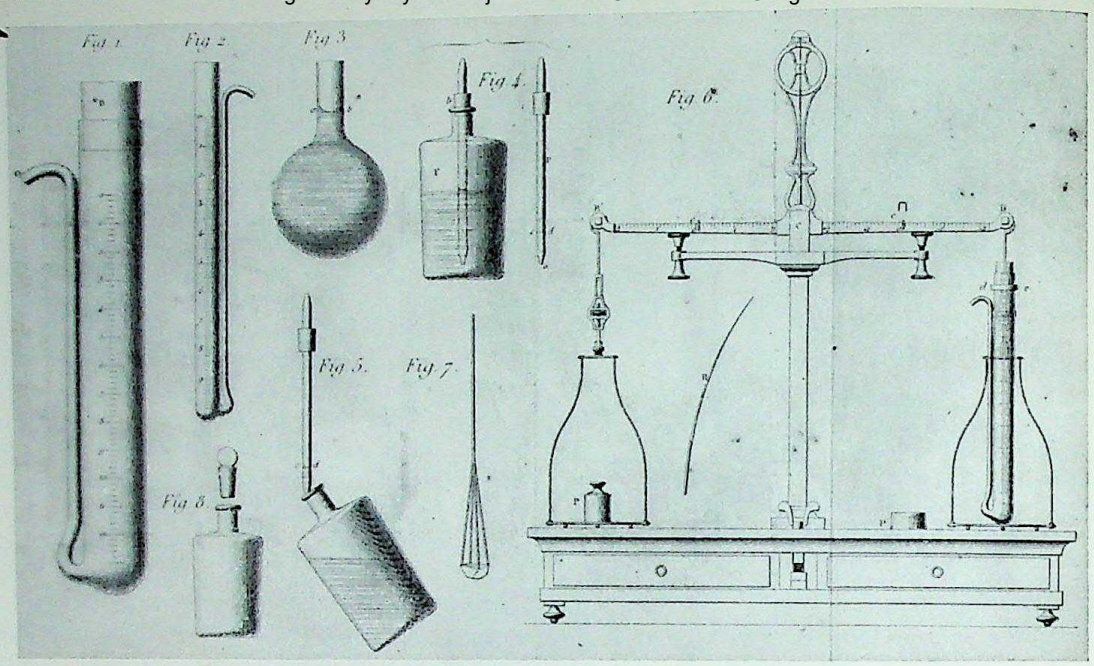

FIGURE 3 – *Apparatus from Gay-Lussac's* Instructions sur l'essai des matières d'argent par la voie humide (1832).

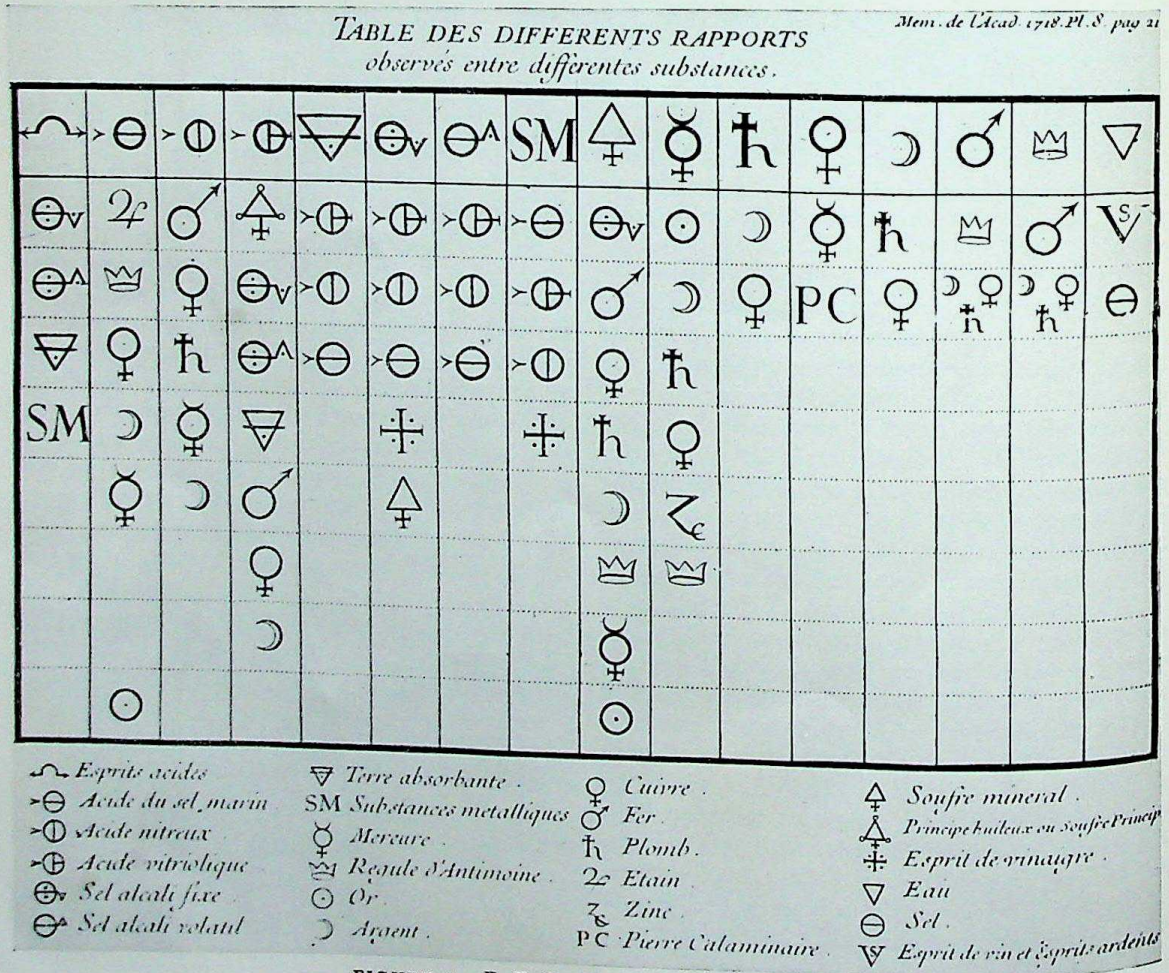

FIGURE 4 – *E. F. Geoffroy's Table of Affinities.*

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a salt solution with the weight of the same volume of pure water. Two hydrometers (figure 2) are also described.

Again, a recurrent problem of medical importance was the nature of mineral waters. As early as the fifteenth century, medical men had made simple tests of the constitution of mineral waters by comparing the appearances of the crystals obtained on evaporation. The iatrochemists of the sixteenth and seventeenth centuries continued to examine mineral waters, but, having no unifying theory, made no useful progress.

Yet another approach used colour changes as tests. Some of these were known from an early date—the nut-gall test for iron was known in antiquity—but their usefulness was naturally limited by the inadequacy of chemical theory. Not until Boyle do we find very much comparison of one test with another. Many of Boyle's reactions are to be found in the pages of other writers, such as Tachenius, but only Boyle seems to have recognized that change of colour, rather than colour itself, might be significant. It is in Boyle that we see one of the salient features of modern chemical analysis—the identification of a substance by inference from some secondary quality developed in particular conditions. Thus, although a number of substances are blue, if the particular conditions of the test—say the solution of a sample of silver in acid—were such that copper was the most likely of the substances, he considered the blue colour as proof of the presence of copper. He understood the value of a succession of tests. One example starts with a test with an indicator to see if a substance is acid or alkaline. Then, if it is alkaline, it can be tested with mercuric chloride solution: a yellow precipitate shows a 'fixed alkali', a white precipitate, a 'fugitive alkali' (ammonia). He even hints at the principle of titrimetry. He points out that an acid discharges the blue colour of tincture of *lignum nephriticum*, and an alkali restores it. 'And I therefore allow myself to guess at the strength of the liquors examined by this experiment, by the quantity of them which is sufficient to destroy or restore the ceruleous colour of our tincture.'

Boyle also devised analytical tests for specific situations. To show that the residue obtained by the prolonged calcination of blood is indistinguishable from sea salt, he concludes a succession of tests thus: some gold leaf is put into aqua fortis. There is no reaction until the blood residue is added. Then the 'liquor which being thereby turned into a kind of aqua regia did in a trice without the assistance of heat totally dissolve it'. He did not found any school of analytical chemistry, but he did at least establish that analytical chemistry must be comparative, rational, and related to general chemical science.

A further step towards a systematization of chemical reactions in a way that could lead to a rational system of analysis was the preparation by E. F. Geoffroy (1672–1731) of his tables of affinities (figure 4). These tried to show the order of affinity of a base for a series of acids, and of an acid for a series of bases. The tables were founded on the assumption that an acid with a higher affinity for a base would displace one with a lower. It was found that these simple tables could not be made to be consistent with one another, but they are important in the present context in that they list members of a limited chemical repertoire which could enter into various combinations while preserving their individual character.

Progress was rapid from the time of T. O. Bergman (1735–84) [12]. He proposed the substitution of analysis in solution for analysis by reactions between dry reagents. One line of investigation was the study of reactions in solution that produced precipitates of known composition. He showed that, by inference from the composition of a precipitate, one could calculate the amount of the species being estimated in the solution of its compound, and he was able to establish a table of compositions that is in effect a table of equivalents. He showed by simple algebra that higher accuracy was to be expected in this way since 'the weight of sediment, from which the quantity of the content is judged is never less, but often greater than that obtained by fire'. His fundamental attitude to the use of precipitation reactions is summed up thus: 'Let us suppose that a quantity of metal (a), precipitated in a certain manner makes a weight (b); if that same manner be exactly employed we may fairly conclude that a quantity of precipitate (nb) occurring in any case is correspondent to a quantity of perfect metal (na), although in the fundamental experiment, either the precipitation is incomplete or some extraneous matter is present'.

This is one of the earliest examples of chemical argument in terms of constancy of composition; this is essential for all analysis not depending, as does assaying, on the isolation of the species sought. It is, of course, a foundation of gravimetric analysis.

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TITRIMETRY

Volumetric analysis was developing concurrently. As E. Rancke Madsen has shown in an admirable study [13], the principles of titrimetry were being worked out early in the eighteenth century. Geoffroy compared concentrations of acetic acid solutions by adding solid potassium carbonate; his end-point was the cessation of effervescence. This kind of acidimetry was often used, but the principle was not generalized until the end of the century. Francis Home (1719–1813) appears to be the first to estimate alkali (potassium carbonate in ash) by titration with an acid (1756). Home's estimates were comparative: William Lewis (1714–81) in 1767 extended the method by standardizing his acid against pure potassium carbonate. Lewis also used litmus paper as an indicator. However, the work of both Home and Lewis was overlooked, and it was only after the work of C. F. Wenzel (1740–93) that colour indicators were widely used.

Another type of reaction that became important was the determination of the relative strengths of bleaching solutions. F. A. H. Descroizilles (1751–1825), in about 1787, described a titrimetric method for the determination of chlorine by indigo, and precipitation reactions, such as the determination of lead carbonate, were also described.

Measurement of the quantities of the titrant delivered had been either by weight, or by crude volumetric methods—Francis Home, for example, used a teaspoon. In 1784 Guyton de Morveau described for the first time a graduated vessel for volumetric determinations. It was what is now called a measuring cylinder, and the reaction took place in it. In 1806 Descroizilles described a burette that delivered solutions from a graduated vessel; it was controlled by a finger over an air-hole near the exit.

Volumetric methods were by then well established, notably in technical chemistry, where they served for estimations where no qualitative problem existed. Gravimetric methods developed more through pure chemistry, where the material handled might be of unknown composition.

The determination of the qualitative composition of an unknown requires a systematic approach depending on some sort of classification. The group system of analysis which formed the basis of the qualitative inorganic analysis that started in the nineteenth century, and is still used, is first clearly recognized in the work of Heinrich Rose (1795–1864), but it had begun to take shape with Bergman [14]. Where the unknown was a mineral, the principal problem was to dissolve it. Bergman ignited minerals with alkali, and extracted with acid. Then followed a series of dichotomic separations by filtration. The residue in each stage was subjected to one treatment, the filtrate to another, resulting in a further suspension which could be separated by filtration, and so on. At the end of each branch some solution or residue would be obtained which could be of only one particular chemical species. However, this system could not be even moderately satisfactory until it had a sound theoretical basis, and this was not available until after the classificatory work of Lavoisier.

The most important stimulus to the co-ordination of analytical chemistry was Dalton's atomic theory. At first sight it would appear as if this theory were the outcome of an array of analytical data, but the converse seems to have been the case: by showing theoretically that atomic weights could be determined from analytical data, Dalton provided an ultimate goal for analysis and a criterion for the excellence of methods. The determination of atomic weights stimulated men like Berzelius to improve greatly on existing methods of analysis, to the benefit of chemistry at large. His pupil Heinrich Rose was able to compile a textbook of the methods; the scheme of the book is recognizably one of the forerunners of systems still used today.

We can sum up the change which had taken place in the climate of analytical chemistry by comparing Tillet's work, as described earlier, with another study that had a similar origin.

Tillet's criticism of the assaying methods of his day had failed to have the effect it might have had because no alternative approach to analysis was commonly available. Seventy years later another criticism of assaying was made, but this time against a background of far more theoretical knowledge.

As before, inconsistency in assay results led to an official inquiry. Alloys of known composition were sent to assayers in several places in France and in other countries. In every case low results were returned. In the course of the general inquiry a committee of technical inquiry was set up, and one member, Gay-Lussac, developed a method in which he had been interested for some time. He published a detailed account in 1832 [1].

In his method the alloy is dissolved in nitric acid and mixed with a solution of marine salt, which precipitates the silver in the form of silver chloride, a compound that is quite insoluble in water and even in acids. The amount of silver chloride

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precipitated is determined, not by its weight, but by the weight or the volume of the solution of marine salt that was necessary exactly to precipitate the dissolved silver.

As he says, the end point is easily recognized by the cessation of all cloudiness as one gradually adds the solution to that of the silver nitrate. He found that one milligram of metal is easily detected in a hundred grams of liquid, and that a half or even a quarter of a milligram can be detected if the liquid is quite clear before the addition of salt solution. Gay-Lussac goes on to describe apparatus for preparing and storing solutions for titration. He describes precautions, and estimates the limits of accuracy of the method.

This is a routine method for a single type of determination, but it would have been impossible for it to be set forward with any confidence unless Gay-Lussac had been sure, firstly, of the constancy of composition of the silver chloride precipitate; belief in constancy of composition is usually considered as originating with Proust, but Bergman had already implied it in his own work on analysis by precipitation. Secondly, Gay-Lussac had to assume that the precipitation resulted from a straightforward double decomposition. This is one of the consequences of the theory of elective affinities of Bergman, a later development of that of Geoffroy. The third assumption was that a single simple substance is precipitated. This is one of the less often noted consequences of Dalton's theory. Although Proust had established the law of constant composition, it did not follow that there was no reason for believing that there might not be more than one substance composed of the same two elements in quantities not rationally related to one another. Dalton's theory does imply such a rational relation, which has the force in this case of implying that the silver chloride formed is likely to be the only compound of silver and chlorine possible.

Fourthly, and this springs also from Dalton and Proust, he assumed that the results obtained are independent of the quantities employed; that is to say, once the specimen has been dissolved, the process yields readings in terms of a ratio or proportion similar to those used in the then current theory of chemical constitution. The 'profitable yield' theory that had sustained the assayers had at last given way to a true theory of chemical constitution.

A great deal of attention is commonly paid in histories of chemistry to the pneumatic chemistry, the gas analysis, of the end of the eighteenth century. Equally important, however, is the rise of analysis by reactions between solutions. The study of the history of analytical chemistry presents incidents as stimulating as any in the history of science; those who apply themselves to it are rewarded by being brought into touch with men whose activities, more so perhaps than those of any other chemists of the past, are immediately intelligible and sympathetic to the chemist of the present day.

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Eastern Africa: the ecological base

SIR JULIAN HUXLEY

In 1960 Sir Julian Huxley visited eastern Africa, at the request of Unesco, to study the conservation of wild life and habitats there. In this article he discusses the ecology of the region and suggests lines on which ecological investigation could best be developed. He also examines the arguments for conserving the wild life of the region and the relationship of such a policy of conservation to traditional methods of agriculture and to planned use of land.

During the Pleistocene, biological evolution reached a climax of efficiency and variety that man has been progressively destroying, eroding, and transforming ever since he became dominant. The most striking ecological climax community of the Pleistocene existed in the drier parts of inter-tropical Africa, on the great tableland that has for many millions of years provided a habitat for very varied flora and fauna. Nowhere else in the tropics—and, of course, nowhere outside the tropics—were conditions so favourable to life.

When I was in eastern Africa in 1960, it was borne in on me that I was privileged to be looking at the last remaining portions of this final, and finest, manifestation of pre-human evolution, which has the merit—not shared by the equally rich rain-forest community—of being readily accessible for observation and study. Its adequate description and analysis have barely begun, and this very urgently needs to be done in the interest of world science [1].

The ecological situation in eastern Africa is broadly as follows. In spite of the abundance of wild game, it is in practice a protein-deficient region. T. A. M. Nash [2], after citing the U.S. National Research Council's statement that the normal human daily intake of protein should be 80 g, of which 30 g should be of animal origin, quotes B. M. Nichol's findings in the tsetse-infested middle belt of Nigeria, which is comparable to eastern Africa. There the total daily intake of protein is 70 g, and of this only 5 g is animal protein [3, 4].

Fisheries, both marine and freshwater, and also fish-culture [36], are very important in the region, both economically and as sources of protein for the local population [5, 6]. Most of the game exists within Africa's huge tsetse-infested area, which extends over some 4½ million square miles of tropical Africa, an area half as large again as the United States [5]. Though native cattle are grazed only on lightly infested areas, many of them get a slight nagana infection (trypanosomiasis) and do not thrive. Apart from this, most of the native domestic stock is kept for prestige reasons and not for meat. East Africa is thus a meat-hungry region, and this encourages illegal slaughter of game, poaching, as it is euphemistically called.

At least a third of the region consists of poor-quality marginal lands. Most of these are ecologically very brittle habitats, liable to deteriorate and lose productivity if cultivated or in the least degree overgrazed. When overgrazing is severe, as in much of the Masai and Samburu areas, the country rapidly degenerates towards desert. Ecological studies by Fraser Darling [12, 13] and by a number of American Fulbright scholars [7–11, 14–16] have shown that many of these marginal habitats produce a considerably larger biomass (mass of animals per unit area) in the form of wild game than they do in the form of domestic stock.

GAME-CROPPING

F. Bourlière, in the 'Wenner-Gren Symposium on African Ecology and Human Evolution (July 1961)' gave the following interesting figures for the biomass of the main wild mammalian species in a number of areas. In a forest area in Ghana, the wild mammalian biomass, even including such creatures as monkeys and squirrels, was less than 0·1 metric ton per km^2.

Lee Talbot finds that, in addition to yielding a

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Area	Authority	Biomass (metric tons per km²)
Ruanda-Ruchuru plain, Parc National Albert, Congo	F. Bourlière and J. Verschuren	24·4
Queen Elizabeth National Park, Uganda	R. Bere	19·5
Nairobi National Park, Kenya	S. I. Ellis	15·6
Serengeti Plains, Tanganyika	B. Grzimek	5·2
Henderson Ranch, S. Rhodesia	R. F. Dassmann and A. S. Mossman	4·9
Kruger National Park, Transvaal	R. Knobel	1·8
Scottish Deer Forest (Red Deer)	V. P. W. Lowe	1·0
Canadian Barren Grounds (Caribou)	A. W. F. Banfield	0·8
Southern Russia (Saiga Antelope)	A. Bannikov	0·35

higher biomass per unit area, wild ungulate populations in eastern Africa have a higher rate of annual increase than domestic cattle. Furthermore, such marginal areas can yield a larger financial return, by way of meat, skins, and other animal products, if managed for game-cropping than if ranched for cattle or thrown open to grazing by African stock. In still other areas the best results are obtained by combining wild game (or domesticated game species such as eland) with domestic cattle. Clearly the study of the ecology of these brittle habitats in general, and of their wild faunal communities in particular, is urgent.

The Republic of South Africa presents a paradox. Its wild game was totally or virtually exterminated except in a few reserves and national parks. But now various authorities, including the municipality of Pretoria, are using parts of their parks or reserves to breed selected wild species, such as blesbok and springbok, and sell numbers of them to farmers in the drier areas, where they continue to breed. The surplus is culled to provide fresh or dried meat and to yield skins and other marketable products. Between 2000 and 3000 farmers are practising this form of game management in the Transvaal alone. Zebra, eland, impala, and other antelopes have proved similarly profitable in Rhodesia.

What is the reason for the superior biological efficiency of wild game over domestic cattle in such circumstances? The answer is that the wild species are part of an ecological community that has reached a high pitch of efficient adaptation through natural selection operating over long periods of time [17]. In a typical eastern African savannah or open-bush habitat, the ecological community is highly differentiated and specialized, as clearly set forth by Fraser Darling [12] in his study of Northern Rhodesian ecology. The animal sector may contain as many as twenty species of herbivore ranging from elephants to tiny dikdik, and including up to fifteen species of antelope [18, 19] each occupying a slightly different niche.

In contrast, domestic cattle are restricted to a few food plants, and so leave much of the habitat unutilized. Furthermore, in the absence of game, some of the plant species not utilized by cattle may increase and so actually reduce the numbers of the species that can be utilized. Cattle are also subject to many diseases, notably nagana, to which indigenous species of ungulates have become immune. Finally, several antelopes have become so well adapted to aridity that they do not merely survive, but actually thrive, on a water ration wholly inadequate for cattle; some species, like the gerenuk, appear never to drink at all.

Consequently, some areas can be made to yield the maximum amount of profit—physiological, financial, and social in various combinations—if they are so managed as to carry an optimum population of wild game species. In some areas, wild-life can be ranched in combination with domestic stock. An annual crop of game animals can be culled and sold at a profit [14].

Such game-cropping schemes are already established on a number of South African farms, and have now been initiated on a few Rhodesian ranches. In Uganda, the protection given to hippos in the Queen Elizabeth National Park led to their over-multiplication, with resultant spread into neighbouring areas. This brought about a far-reaching destruction of their habitat within and near the Park. Experimental culling in selected areas proved effective in restoring the habitat, and hippo cropping is now being continued, both within the Park and, on a larger and

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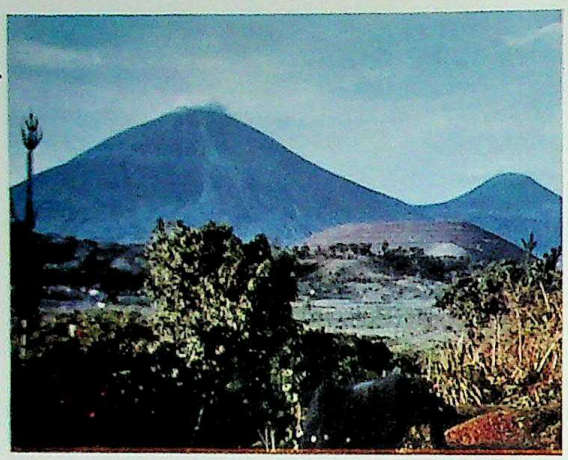

FIGURE 1 – *Mount Muhavura and Mount Mgahinga, part of the Virunga volcanoes, on the Uganda–Congo border. Mountain gorillas live in a reserve on their slopes.*

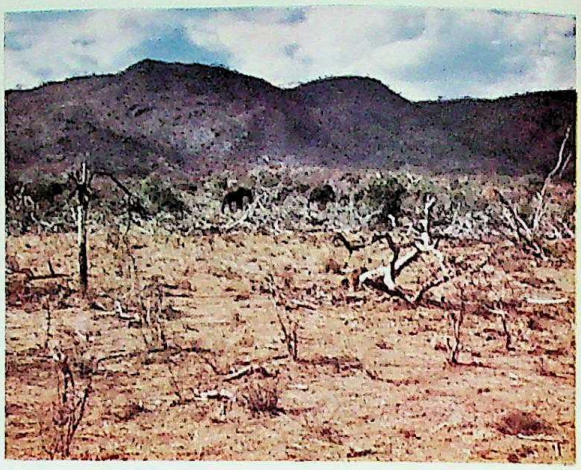

FIGURE 2 – *Elephants among the devastation they have caused in the Tsavo National Park, Kenya.*

FIGURE 3 – *The Teleki volcano at the south end of Lake Rudolf, with a lava flow beside it.*

FIGURE 4 – *A buffalo skull with horns infested with a species of Tinea, a close relative of the common clothes-moth.*

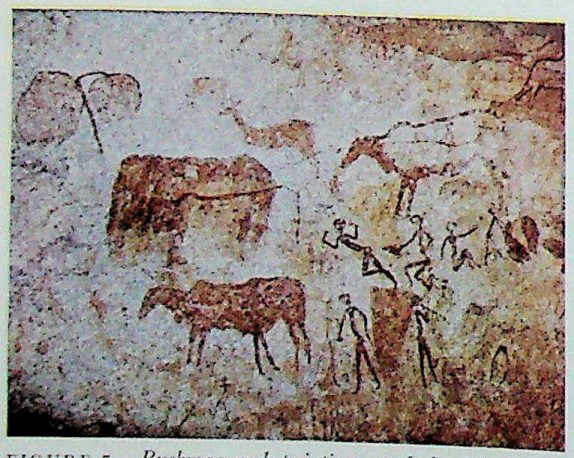

FIGURE 5 – *Bushman rock-paintings at Lake McIlwaine, Southern Rhodesia, showing bushman hunters, eland and other game species, and a curious tree.*

FIGURE 6 – *A rescue team with their boat at the edge of a sheet of* Salvinia *on Lake Kariba.*

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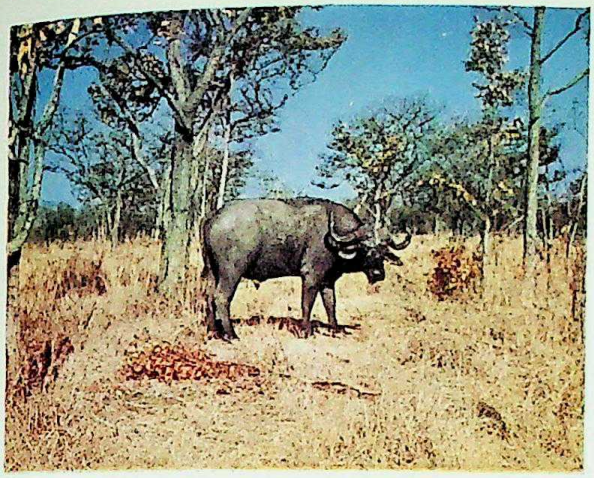

FIGURE 7 – *A lone buffalo bull by the roadside in the Hluhluwe Reserve, Natal.*

FIGURE 8 – *A mother hippo and her baby near the Murchison Falls, National Park, Uganda.*

FIGURE 9 – *A bush-fire, Lochinvar Ranch, Northern Rhodesia.*

FIGURE 10 – *A pride of lions finishing a zebra carcass, Gorongoza National Park, Mozambique.*

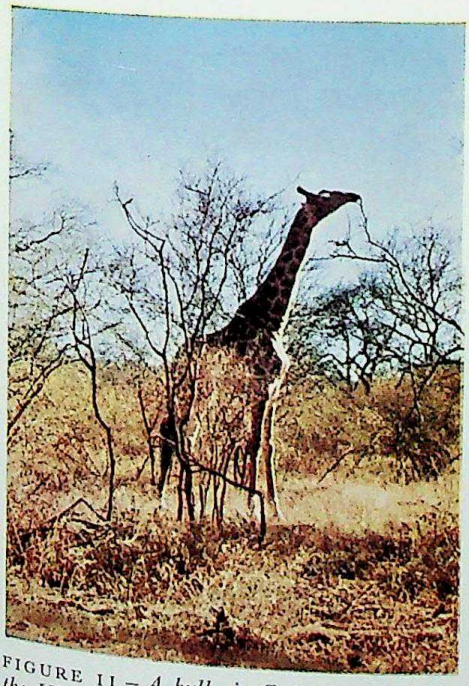

FIGURE 11 – *A bull giraffe browsing high in the Hluhluwe Reserve, Natal.*

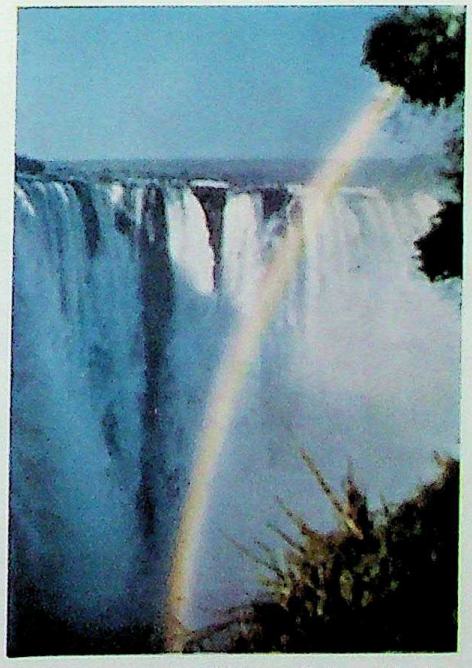

FIGURE 12 – *The Victoria Falls with their rainbow, seen across the western end of the gorge from the south, Natal.*

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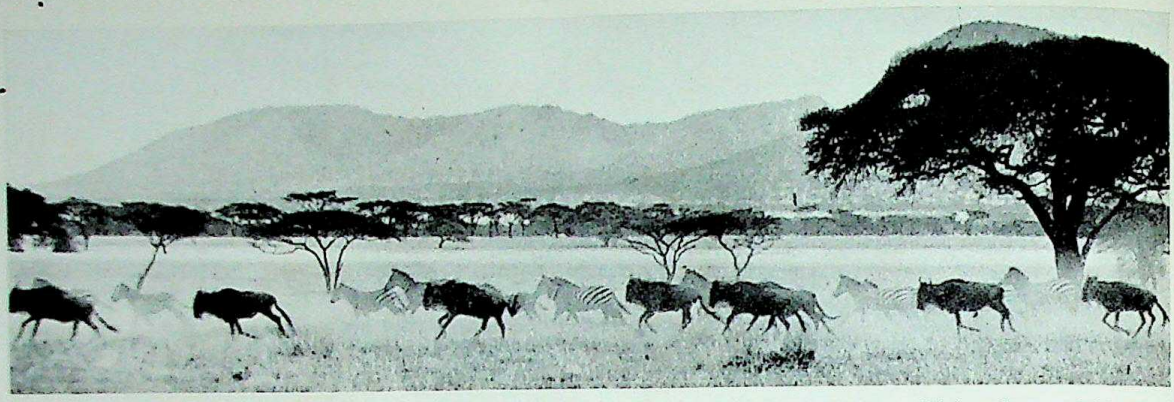

FIGURE 13 – *A mixed herd of zebra and wildebeest galloping past a fine acacia on the Seronera Plains, Serengeti National Park, Tanganyika.*

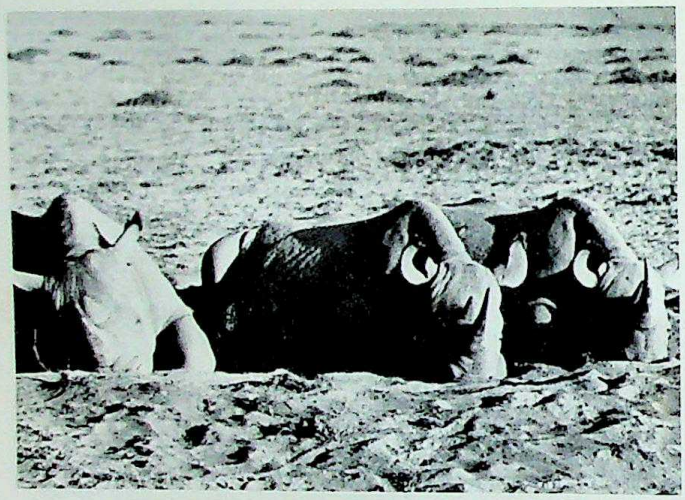

FIGURE 14 – *Three young black rhinos taking a dust-bath in the Amboseli Tribal Park, Kenya.*

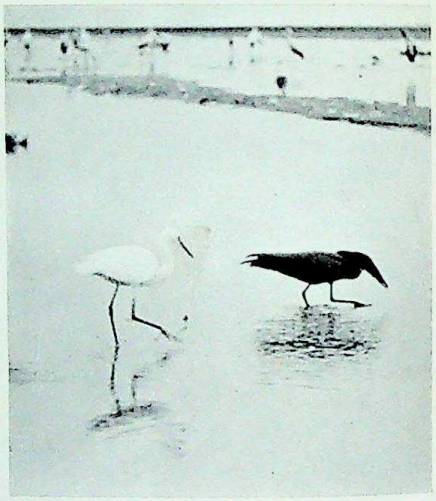

FIGURE 15 – *An egret and a hammerkop* (Scops umbretta) *in Lake Manyara National Park, Tanganyika.*

FIGURE 16 – *An African fish eagle taking flight, Murchison Falls, National Park.*

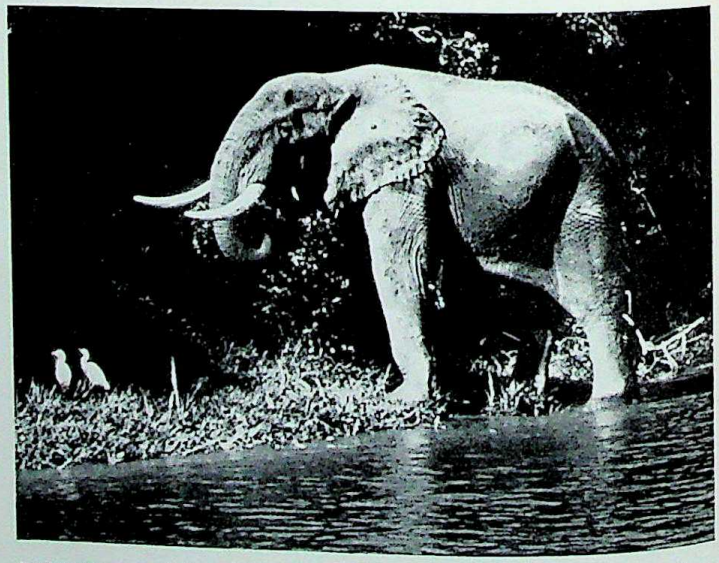

FIGURE 17 – *A lone bull elephant with some cattle egrets on the Kazinga Channel, Queen Elizabeth National Park, Uganda.*

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more organized scale, in areas just outside it [9] (figure 8). As a result, some 5000 tons of hippo meat is now being made available annually.

With the support of the Nuffield Foundation, an even more interesting scheme has been initiated in the Galana River area of the Waliangulu tribe, adjacent to the Tsavo National Park in Kenya. The elephants in the Park have seriously over-multiplied, damaging and killing numbers of baobab and other trees (figure 2) and spreading into the Galana River region. Selected hunters from the Waliangulu, traditionally a hunting tribe, are being appointed to kill the surplus elephants. The proceeds of the operation will go to the tribe, either as food, or as profit from the sale of meat, ivory, and other products. A similar project, but aimed at a mixed crop of various bush species, is being put into operation in a region adjacent to the Meru African District Park in Kenya, and others are planned.

In principle, game-cropping should contribute materially to the solution of various eastern African problems, but if it is to be effective, a great deal of research and study will be needed. It is necessary to know how to preserve the meat; how best to transport and market it and other products; whom to train to do the actual culling. It is necessary also to ensure that the local communities shall not only reap the fullest benefit from such schemes, but shall understand their value, for without this their co-operation will not be obtainable.

POACHING

This last consideration raises the problem of poaching, for no amount of education alone will abolish this hateful business [18]. Poaching in eastern Africa is not a matter of individual Africans setting out to kill a buck or two for their own use; it is a highly profitable, and often highly organized, crime. In Northern Rhodesia poaching gangs supply the mining townships with illegally killed game meat. Large areas are being devastated by fires deliberately lit by the poachers to drive the game to its death in pits or snares. Gangs armed with poisoned arrows watch the water-holes. In some areas the killing of crocodiles for their skins has greatly reduced, or even exterminated, the species [20]. Poaching in eastern Africa today is one of the most serious manifestations of social pathology. Its cure depends on a new pattern of biological and social ecology for the region.

The main source of profit to poachers from the tens of thousands of animals they kill annually in eastern Africa today is not meat but other products, such as skins, sinews, tusks, and horns. The story of rhino horn is the strangest and the saddest. Although its aphrodisiac properties are wholly mythical, belief in them is so strong that it fetches huge prices. Rhino are protected, but it is remarkable how many are killed in 'self-defence.' Ivory fetches much lower prices, but there is much more of it—a pair of tusks weighs between 50 and 100 lb, as against 7 or 8 lb for a rhino horn. It is estimated that the total of rhino killed annually in Kenya alone is 700–900, and of elephants 4000–5000. Though East African elephants seem able to hold their own in a remarkable way, the much smaller and less biologically resourceful rhino population cannot possibly support such a drain for more than a few decades at most.

LAND-USE PLANNING

Eastern Africa still has the chance of planning its future scientifically, that is, ecologically. If it is not to lose this chance, it must speedily make a general land-use survey, as a basis for land-use policy and planning. Presumably this will have to be undertaken by the different territories and governments separately, but it will be a great advantage if the various surveys can be co-ordinated on the basis of some common plans and principles. A prerequisite for such plans is to get rid of the assumption that agricultural or other artificial development of natural habitats is necessarily always good and inherently desirable. It is natural for Europeans and other inhabitants of temperate lands with good soils to make this assumption, and it certainly applies to favoured areas of the region, such as the Kenya Highlands. But it is not true for the marginal habitats of eastern Africa, or for most of the equatorial rain-forest area [12, 13, 21–24].

The primary purpose of such land-use surveys would be to define three main categories of land. Firstly, land to be allocated to development, including agriculture, industrial uses, and urban development. Secondly, land to be conserved and managed as wild habitat, either for game-cropping schemes; for game-viewing in parks and reserves; for soil conservation and watershed protection, as in certain types of forest; for timber as in other forests; or simply to be kept undeveloped in reserve. Thirdly, land in which the aim is a profitable symbiosis of wild and domestic stock, as in areas of African pastoralism like Masailand.

In the first category of lands, wild animals are in general destructive, and should be either kept

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down or—in the case of certain species, like lions, rhinos, and elephants—exterminated or removed. In the second category, the wild community should be conserved, and the wild habitat scientifically managed. In the third category, a balance between wild and domestic species should be established: the primary aim here will be to educate the African pastoralists and to devise schemes for which they will have responsibility and from which they will derive benefit. In Tanganyika, W. H. Pearsall's remarkable ecological survey of the Serengeti (1957) has led to the establishment of a special Conservation Authority in the region.

Already a beginning is being made with such plans. In Kenya, the responsible Minister last year outlined a wild-life policy that will operate very much along these lines. Northern Rhodesia has issued a White Paper on its wild-life policy. Southern Rhodesia is considering a scheme of land apportionment in which there will be a category of national lands which will include not only forest reserves, game reserves, and national parks, but areas to be kept as wild habitats. These will not be opened up for cultivation or ranching until the best use for them (which may well turn out to be for wild-life management and game-cropping) has been determined.

In Northern Rhodesia, the basis for a general plan has been laid by Fraser Darling's reconnaissance [12]. A survey is now being made of the Luangwa Valley, and of the entire Kafue River Basin, though in the latter case apparently without due attention being given to wild life as a resource and to ecology in general. It is to be hoped that such surveys will be continued elsewhere, and that there will be as much regional co-ordination as possible.

ECOLOGICAL RESEARCH

Pari passu with land-use surveys, intensive large-scale ecological research is urgently needed. Pure research is needed on the wild-life communities, especially on the large terrestrial mammals. In particular, the African antelopes provide one of the most striking examples of adaptive radiation [18, 19], and they should be carefully studied from the point of view of evolutionary ecology.

Between East Africa and the Cape region there is an interesting gradation in the proportion of tropical and temperate species. This holds for many animal groups, including mammals, birds, and amphibians, and for many plant groups also. The temperate flora and fauna are the more ancient; the tropical groups are mostly more recent immigrants, which have increasingly pressed southwards with the climatic amelioration since the last glaciation. It will be of great interest to make a comparative study of subspeciation in the region, both from the point of view of the groups concerned and in relation to habitat and environmental changes.

A curious ecological problem has arisen from the creation of the Kariba Lake; this is the largest artificial lake in the world, nearly two hundred miles long. An introduced species of the floating water-fern *Salvinia*, which in the open Zambesi was confined to small areas of still water, has spread as a green carpet over the surface of the shallow bays of the lake to cover more than 200 square miles. This carpet (figure 6), so dense that it can support a prone man, is suffocating the fish population by preventing access of oxygen [25, 26]. Large-scale research is now being undertaken to rid the lake of this menace.

Recently an important new step has been taken in studying the ecology of eastern Africa. This is the setting up of the Nuffield Unit of Tropical Animal Ecology, in which the zoology department of the University of Cambridge, Makerere College, Kampala, and the National Parks Authority of Uganda are co-operating. It will begin with a study of the ecology of the larger terrestrial herbivores of Uganda. It is much to be hoped that similar schemes will be undertaken elsewhere.

Isolated studies have already been made of the territorial behaviour of Uganda kob [27], of the ecology of Thomson's gazelle [28], of the total game population of the Serengeti and its migration routes, of the effect of reducing excessive population-density in hippos, of the reproductive behaviour of giraffes, of the ecology of the lechwe [1], and so on. But this is only a beginning. In order to understand eastern African mammalian ecology as a basis for intelligent practical action, we need, as a minimum, an organized study of reproduction and development, longevity and population genetics, ethology and habits, nutrition and food-preferences, migration, and interactions with other species, in all the main mammalian members—say twenty to twenty-five species—of eastern African communities. As a basis for such work, we urgently need a reliable handbook of African mammals, coupled with a series of good field-guides.

In order to understand the general ecology of the region, we must study other groups than mammals. A good beginning has been made with

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Africa's splendid avifauna. But, as Fraser Darling writes [12], 'a great world of discovery remains in Africa in determining the role of soil invertebrates in the conversion cycle: they are just as important as the large animals above ground'. He cites a species of millipede as playing a key part in utilizing the detritus of an important grass species; and termites, of course, play an outstanding role.

Some species of African animals are, or are said to be, in real danger of extinction—either total extinction, or extinction everywhere except in small protected areas. I may instance the mountain gorilla, both the white and the black rhinoceros (figure 14), and the bongo (*Boocercus* spp.). The Survival Service of the International Union for the Conservation of Nature is already doing valuable work in collecting facts about such species and bringing them to public notice [29], but detailed ecological research and survey are needed in every case. The value of such study is shown by the work of J. T. Emlen and G. B. Schaller [30], who have shown that the intensely interesting mountain gorilla (*Gorilla g. beringei*) is not, as has been suggested, in imminent danger of extinction; between 5000 and 11 000 still exist, in a number of habitats (figure 1) in Western Uganda and the eastern Congo. For large-scale survey, air photography is essential. This can also be useful in animal census work, though it should be combined with aerial counting and ground sampling [31].

Immobilizing drugs have been extensively studied. These are important, firstly to enable selected wild animals to be taken for study and measurement, and tagged or marked for future identification; and secondly to transport wild animals from dangerous situations to safety. The drugs are injected from specially designed ampoules, usually propelled by CO_2 under pressure, and discharged from some kind of weapon [32].

With some large tough-hided species, the use of such drugs is difficult and even dangerous, and here one must rely on skill and courage to lasso, tie up, and transport the animals. A recent successful use of this method may be mentioned. The rare white rhinoceros—which is the third-largest terrestrial animal in existence, and is specialized for grazing instead of for browsing like the black rhino—today has a peculiar discontinuous distribution in two areas. One is in Natal, the other in north-western Uganda and the southern Sudan, on the left bank of the Albert Nile; the river has proved an insuperable barrier to the easterly spread of the species. In the Uganda portion of this area there is no national park or adequate game reserve, and the very survival of the animal is threatened. It is known that the white and black rhino can coexist happily in the same area, and accordingly, after some preliminary ecological studies, a number of white rhinos were captured, transported across the Nile, and released in the Murchison Falls National Park, where they appear to be flourishing.

The most important pathological significance of wild game is that many species harbour large numbers of trypanosomes in their blood without developing any disease-symptoms; they can therefore act as reservoirs for infecting domestic stock with nagana. As is well known, various governments have carried out wholesale slaughter of game with the aim of making large areas safe for cattle; in Southern Rhodesia alone, over half a million wild animals have been killed. The last annual report on the subject announced that a record number (36 552) had been killed in a year, the majority ungulates, but including also lions, leopards, elephants, and rhinos [1, 33]. It is now clear that this policy has little scientific basis. Research has shown that a number of species which cannot be eradicated in this way—various small mammals, and even some reptiles—are also attacked by tsetse and may harbour the parasites of nagana. As a result, the policy of game slaughter has now been abandoned, except for local programmes where it is combined with bush clearance followed by agricultural settlement.

Thanks to research, nagana disease in cattle can now be successfully prevented, either by injecting the animals with anti-trypanosome drugs such as Antrycide, or by selective bush clearance leading to the disappearance of the tsetse vector. It is well to remember, however, that this scientific success poses further practical problems. Thus both clearance and anti-trypanosome injections are expensive, and must be repeated at intervals. The question thus arises whether it will be more profitable—financially, physiologically, or socially—to get rid of the tsetse or the active trypanosomes and introduce cattle, or to manage the habitat for the production of protein by game-cropping. In some cases, would not the introduction of native cattle inevitably or probably lead to the loss of the wild habitat's productivity, and to its progressive deterioration towards desert? Only new research or scientific survey based on established knowledge can provide the answers to such questions.

The effects of fire (figure 9) also need much long-term research [1, 12]. No one who has not travelled in eastern Africa can realize the amount

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of burning that goes on. At night it is rare not to see one or more lines of flame lighting up the dark horizon: flying over the country by day, one sees innumerable blackened patches, large and small. Natural fires, caused by lightning, have always occurred in Africa, but with the coming of man, deliberate burning was undertaken, either to clear land for shifting cultivation, to secure the growth of fresh grass, to smoke out wild bees' nests for honey, or to drive game in a desired direction. All such fires may get out of hand.

Burning is probably necessary for the proper maintenance of the wild habitat today: certainly all Game Departments and National Park Authorities employ burning as part of their system of management. But there is much disagreement as to whether early or late burning is better, and whether bush should be burnt annually, or every two or every three years. The answers will probably differ in different areas, but only painstaking experimentation can decide what is best.

Another field where much more research is needed is in ethology—the study of the behaviour of animals—notably perhaps that of especially interesting species such as elephant, rhinoceros, gorilla, and chimpanzee. Special anthropoid observatories—both for continuous investigations, and for short-term studies by visiting scientists—might well be built in suitable areas. It would be of special interest to devise methods for the better observation of nocturnal species, including such strange and little-studied forms as aardvark and aardwolf. Observatories or large hides on the edge of lakes or swamps would provide fine opportunities for the bird ethologist.

THE ENJOYMENT OF WILD LIFE

The sight of an abundance of large wild animals in freedom is an unforgettable experience, and people will travel long distances to enjoy it. The tourist trade is therefore of considerable and rapidly growing economic importance to eastern Africa. The proper utilization of this resource demands the creation of national parks and reserves in which adequate samples of the wild life of the area can be preserved. Essential requirements are good access, accommodation, and viewing facilities [see 1]. Surveys should be undertaken to determine which areas deserve protection for their scientific, scenic, or historic importance and which deserve the status of national or tribal parks.

In Britain, on the recommendation of the National Parks Committee, an official list has been prepared of areas representing all the main natural or semi-natural habitats, and also the main geological formations and features. A new statutory body, the Nature Conservancy, was set up with the duty of acquiring these as opportunity offered and of administering them as National Nature Reserves. Similar lists should be prepared for all eastern African territories. In my view they should include not only a complete sample of habitats and geological formations, but also archaeological sites, historic monuments, and places of outstanding natural beauty. Examples of sites that should figure in such a list are the famous stone buildings of Zimbabwe and elsewhere; the huge earthwork of Iron Age type in eastern Uganda; various features of the great African Rift system; Leakey's remarkable excavations at Olduvai, which have revealed the history of man and life throughout the Pleistocene [34, 35]; numerous sites with prehistoric and historic rock-paintings (figure 5); giant calderas like Ngorongoro and active volcanoes like Ol Doinyo Lengai and Nyamlagira; beauty spots like Lake Bunyoni; and historical monuments like the slavers' coastal headquarters.

A comprehensive system of national parks should also be set up; but for these the criteria are rather more complex. Firstly, the national parks must be numerous enough to include all major types of suitable country, not merely the game-rich savannahs. Secondly, the high prestige value of national parks must be balanced against the equally unquestionable land-hunger of most African peoples. Thirdly, the designation and establishment of tribal parks must be undertaken in the light of careful socio-political study. Masailand is the most conspicuous and most important example. Straddling Kenya and Tanganyika, it contains the largest and most varied reservoir of spectacular wild mammals in the world. Much of it is being badly overgrazed by herds of poor, but nevertheless rapidly multiplying, native stock, and the Masai are beginning to regard the game animals as competitors for the grazing. The aim should be to establish a symbiotic relationship between the game, the domestic stock, and the Masai, from which all would benefit. The ways of achieving this should include making over to the Masai as much as possible of the financial benefits from hunting licenses, game meat, found ivory, national parks, and tourism in the area. Masai tribal parks should be established to attract visitors, with adjacent areas for game-cropping; these would act as a source not only of

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revenue, but of political prestige. In July 1961 it was announced that the famous Amboseli Reserve in southern Kenya had been made over to the Masai as a tribal park of this kind. It will be interesting to see how the necessary scientific and ecological control will be maintained.

ECOLOGY IN AFRICAN EDUCATION

Ecology is also of great importance in African education. In this context I am not thinking solely of the problem, urgent though it is, of educating African public opinion to regard wild life as a major resource. I want to stress the need for a biological, and notably an ecological, approach in school and college education at all levels. In a region like eastern Africa, the best way of introducing boys or girls to science is, I believe, not through physics and chemistry but through the earth and life sciences, above all ecology. Such an approach readily leads on to analysis in physico-chemical terms, and it also links up with physiology—notably nutrition and reproduction—and so with human ecology and the whole problem of resource conservation. I regard the working out of a scientific curriculum adapted to the needs of underdeveloped tropical countries like Africa as one of the urgent tasks awaiting Unesco and all professional educational bodies.

Technical training for those professionally concerned with conservation, and general courses on conservation and land-use for administrators and others whose work impinges on human ecology, are also essential. A beginning has recently been made with courses of the former type at the University College of Rhodesia and Nyasaland in Salisbury, and with those of the latter type at University College, London.

The problems of eastern Africa can keep the world's natural and social scientists busy for a long time to come. In particular, their challenge will undoubtedly prove a powerful stimulus to the development of ecology, both biological and human.

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Masers and lasers

R. A. SMITH

It has been found that microwaves will stimulate a substance in a particular energy-state to add to the microwave beam. This microwave amplification by stimulated emission of radiation, known, from its initials, as maser action, is almost completely free of 'noise'. This has made it possible to build extremely sensitive microwave amplifiers. An analogous device for light-waves, the laser, produces a very narrow beam of high energy. This article discusses both the maser and the laser and the theory on which they rest.

STIMULATED EMISSION OF RADIATION

It is amazing how many scientific developments have sprung from ideas originated by Einstein, although, to all except professional physicists, he is most generally known for his work on the theory of relativity. The fundamental ideas on which the developments discussed in this article are based were introduced by Einstein in his discussion of the acute problems that arose in the early days of the quantum theory of radiation. When an atom is in an excited state, that is a state of higher energy than its normal, 'ground', state, it emits radiation by dropping to a state of lower energy. This was generally considered to be a random process in which each type of atom has an average lifetime in the excited state. What Einstein proposed was that, in addition to this random process, the descent from an excited state could be initiated by radiation of appropriate frequency; this type of emission is known as stimulated emission. He showed, moreover, that the probability of this process taking place is proportional to the intensity of the radiation field in which the atom is situated, and is exactly equal to the probability of excitation of an atom from the lower state to the higher state by the radiation. There is just one process for excitation by radiation, the absorption of a quantum of radiation; for de-excitation there are two processes, stimulated emission of a quantum and spontaneous emission of a quantum. The latter is generally by far the more important and the former has frequently been forgotten. However, as we shall see, in certain circumstances stimulated emission may predominate. The three processes are illustrated in figure 1.

ENERGY RELATIONSHIPS

In order to discuss the relative importance of stimulated emission and spontaneous emission, we must first of all recall certain simple relationships between the energy of the atomic states and the frequency of their radiation. According to the quantum theory, an atom can exist only in certain states having well-defined energies, which we shall label as $E_1, E_2, E_3 \ldots$ in ascending order of magnitude. The lowest, or 'ground', state will be that having energy E_1, and when the atom is not in this state we say that it is 'excited'. When radiation falls on an atom, it is absorbed in only narrow 'lines' of frequency ν_{21} given by Planck's well-known relationship

$$h\nu_{21} = E_2 - E_1 \qquad \ldots (1)$$

Ideally, these absorption lines would be infinitely sharp, but there are various processes that broaden them into narrow bands; such processes are the Doppler shifts due to the atom's motion, and the variation in the energy of the atom necessitated by Heisenberg's Uncertainty Principle. For the moment, however, we shall ignore this broadening of the lines. The emission spectrum corresponds to the absorption spectrum; that is, the frequencies of its lines are also given by equation (1).

When a large number of atoms are in thermal equilibrium, they will be in a radiation field corresponding to 'black-body' radiation, that is, the intensity of the radiation depends only on the absolute temperature, T, at which the equilibrium is established. Under these conditions, the populations of all allowed energy levels are determined by statistical laws and depend only on the

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Was born in Scotland and was educated at Edinburgh and Cambridge. After research in the Cavendish Laboratory, he held academic posts at St Andrews and Reading Universities before the war. During the war he worked on radar, and in 1946 became Head of the Physics Department at the Royal Radar Establishment, Malvern, a post which he held until recently. He is now Professor of Physics at Sheffield University. He has published a number of books on radar, and on infra-red and semiconductor physics, the fields in which most of his research has been carried out.

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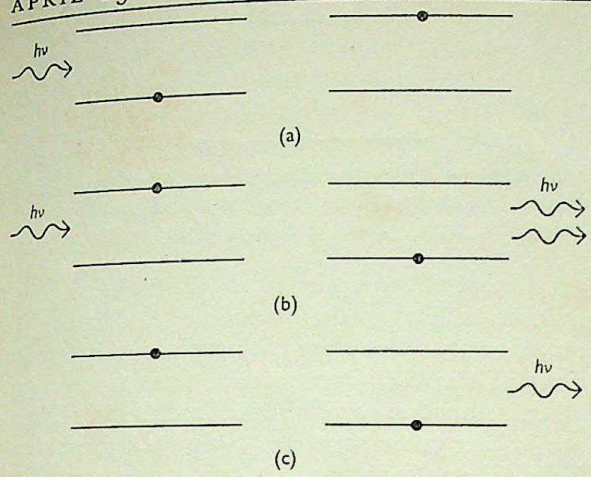

FIGURE 1 – *Illustrating the three processes of absorption and emission of radiation.* (a) *absorption,* (b) *stimulated emission,* (c) *spontaneous emission. Left part of diagram shows the state before interaction; the right, that after.*

temperature. We shall later write down the equations that give the relative populations, but we may say that the higher the energy, E_r, the smaller the number of atoms with the corresponding energy (see figure 2). Suppose, for simplicity, we consider only two levels with energies E_1, E_2 ($E_1 < E_2$). Then if $(E_2 - E_1)$ is large (say of the order of an electron-volt), then at ordinary temperatures almost all the atoms will be in the level with energy E_1 and very few in the other. On the other hand, if E_1 is almost equal to E_2 (say $E_2 - E_1$ is about 10^{-4} eV, corresponding to a transition in the microwave region of the spectrum), then the numbers of atoms with energies in the two levels at ordinary temperatures will be nearly equal. The number in the higher level will be only slightly less than the number in the lower level, and one must work at very low temperatures to obtain an appreciable difference in population.

Let us now state these properties in a quantitative way. If $\mathcal{N}(E_r)$ is the average number of atoms on the level with energy E_r, then we have

$$\mathcal{N}(E_r) = \mathcal{N}(E_1) \exp[-(E_r - E_1)/kT] \quad \ldots (2)$$

where k is Boltzmann's constant; this is known as Boltzmann's law. Since $E_r > E_1$, we see that the number decreases as E_r increases (see figure 2). If $(E_2 - E_1)$ is much greater than kT, then almost all the atoms will be in the ground state. If, however, E_2 is nearly equal to E_1 so that $(E_2 - E_1) \ll kT$ and all the other levels are much farther away, then the atoms are almost equally shared between levels 1 and 2. We may express equation 2 for the populations of the levels with energies E_1 and E_2:

$$\mathcal{N}(E_2)/\mathcal{N}(E_1) = \exp[-(E_2 - E_1)/kT] \quad \ldots (3)$$

Using equation 1 for the frequency of the emission or absorption line associated with the levels, we may also express this relationship in the form:

$$\mathcal{N}(E_2)/\mathcal{N}(E_1) = \exp[-h\nu/kT] \quad \ldots (4)$$

where ν is the frequency of the emission or absorption line associated with the two levels.

Let us now consider some numerical values. If ν corresponds to green light, the energy $h\nu$ has a value of about 2 eV. At room temperature ($T \simeq 300°$K), kT has the approximate value of 1/40 eV, so that $h\nu/kT \simeq 80$. Thus $\mathcal{N}_2 \ll \mathcal{N}_1$ and almost all the atoms, at equilibrium, will be in the ground state. In considering optical absorption, then, stimulated emission may be entirely ignored except at very high temperatures, such as occur in the interiors of stars. If, however, ν is a microwave frequency, corresponding to a wavelength of 10 cm, then $h\nu \simeq 10^{-5}$ eV, and at room temperature $h\nu/kT \simeq 4 \times 10^{-4}$, so that $h\nu \ll kT$. $\mathcal{N}_1$ is then almost equal to $\mathcal{N}_2$, and since we may write in this case $e^x \simeq 1 + x$, we have

$$\mathcal{N}_1 - \mathcal{N}_2 = \mathcal{N}h\nu/2kT \quad \ldots (5)$$

where $\mathcal{N} \simeq \mathcal{N}_1 + \mathcal{N}_2$ is the total number of atoms under consideration.

Even at the temperature of liquid helium boiling under atmospheric pressure (4° K) we should have $\mathcal{N}_1 - \mathcal{N}_2 = \mathcal{N}/50$, so that $\mathcal{N}_1$ is still approximately equal to $\mathcal{N}_2$.

RELATIVE IMPORTANCE OF SPONTANEOUS AND STIMULATED EMISSION

We must now return to consider the relative importance of the two processes of de-excitation, spontaneous emission and stimulated emission. Their relative importance will depend on the

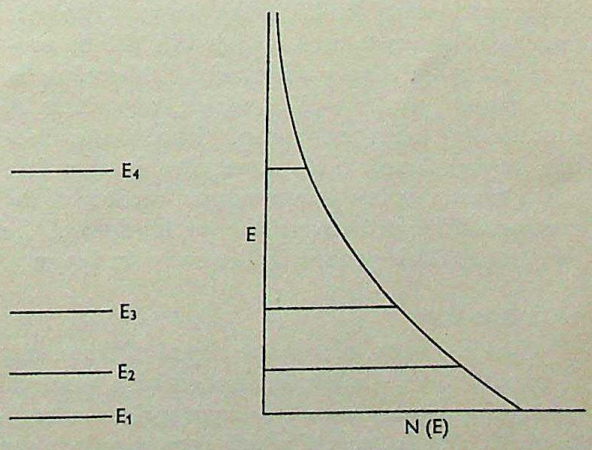

FIGURE 2 – *Variation with the energy* E *of the population* N(E) *of energy levels in thermal equilibrium.*

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intensity of the radiation field, but in conditions of equilibrium the ratio of the probability of spontaneous emission to that of stimulated emission may readily be shown [1] to be $\exp(h\nu/kT)-1$. Thus when $h\nu \gg kT$, spontaneous emission is very much more probable and stimulated emission, in most circumstances, may be neglected. This is why emission of γ-rays from atomic nuclei and emission of visible light from excited atoms are generally regarded as completely random processes. While the former may always be regarded as such, we shall see later that, even for visible light, stimulated emission under certain circumstances can be of importance. On the other hand, when $h\nu \ll kT$, as in the microwave region of the spectrum, the ratio of the probabilities of spontaneous to stimulated emission is approximately equal to $h\nu/kT$ and is small. In this case it is spontaneous emission that may be neglected.

WAVE ABSORPTION

Let us consider the conditions under which absorption will occur. If the populations of the two levels N_1, N_2 were exactly equal, there would be no detectable absorption, since, according to Einstein, the probabilities of excitation and de-excitation for any particular pair of levels would be exactly equal. Thus for every atom excited to a higher level, one would be de-excited to the lower level. It is in fact only the difference between the populations N_1 and N_2 that results in absorption. The number of atoms excited to the higher level will clearly be proportional to N_1, the number in the lower level; the number de-excited (neglecting spontaneous emission) will be proportional to N_2, the number in the higher level, and the constant of proportionality will be the same. The net number of atoms excited, which will also be proportional to the energy absorbed, will therefore be proportional to (N_1-N_2), which, according to equation (5), is inversely proportional to the absolute temperature. The absorption is therefore much greater at low temperatures, because of the greater difference between the populations of the two levels. This process is illustrated in figure 3.

SATURATION

When a strong microwave signal of frequency ν, equal to $(E_2-E_1)/h$, is applied to a material containing atoms with two appropriate energy-levels E_1 and E_2, one of which corresponds to the 'ground' state, absorption will take place, and more atoms will be excited than de-excited. The

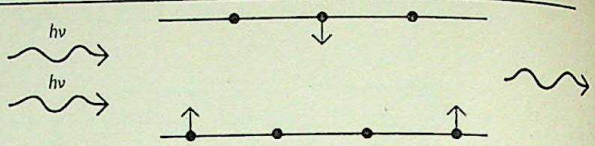

FIGURE 3 – *Microwave absorption. (Here two incident quanta are absorbed and one quantum is emitted through stimulation, leaving a net absorption of one quantum.)*

population of the upper level will therefore begin to increase, that of the lower level to decrease. If no other processes were taking place, the populations of the two levels would tend to equality, when absorption would be reduced to zero. There are in fact other processes taking place, particularly if the active atoms are located in a crystalline solid, as is frequently the case, and these tend to restore populations to the equilibrium value. For example, the excited atom may transfer its energy of excitation to the crystalline lattice. In these conditions the populations would return, if left undisturbed, to their equilibrium values, in a relaxation time τ. Provided τ is sufficiently long, that is, if the atoms are relatively loosely coupled together, the populations of the two levels may still be made nearly equal by means of a large microwave signal of the correct frequency. This process is called saturation. Generally, if the active atoms are in solids, they have to be included in dilute concentration in a host crystal so as to give a sufficiently large value of τ. For example, closely spaced levels are frequently obtained by using paramagnetic ions in a magnetic field; the paramagnetic ions are spread throughout a diamagnetic crystal. An example is the use of Cr^{3+} ions dispersed in alumina. This process is of great practical importance, as will be seen in the following discussion.

MICROWAVE AMPLIFICATION BY STIMULATED EMISSION—MASER ACTION

We have seen that, in conditions of thermal equilibrium, the population of the upper level, N_2, is always less than that of the lower level, N_1, even if only slightly so. By means of the process of saturation we can make $N_1=N_2$ and reduce the absorption to zero. Is it possible to go further, to make N_2 greater than N_1? If so, amplification would occur, since more atoms would be de-excited from the upper level than excited from the lower level. Thus, instead of quanta being taken from the incident radiation, quanta would be added to it. Moreover, this is a true amplification and not just addition of random quanta. The process of absorption is generally realized to be a

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coherent process; a sinusoidal wave-train has its amplitude diminished but retains its phase coherence. The process of stimulated emission is essentially this process reversed, so that a sinusoidal wave grows in amplitude and retains its phase coherence. Generation of radiation by spontaneous emission, on the other hand, is essentially a random process, so that such emission has no phase coherence—it is just 'noise'. This is perhaps the most important distinction between the two processes.

Is it actually possible to make the population of the upper level greater than that of the lower, that is, 'invert the populations' of the two levels? The answer is that it can be done only by human intervention, since, in equilibrium conditions, we always have the population of the lower level greater. That the inversion of the populations can be achieved practically was first shown in 1955 [2], and the developments discussed in this article are a consequence. This process is known as maser action, from the phrase Microwave Amplification through Stimulated Emission of Radiation; amplifiers and oscillators based on the process are called masers.

METHODS OF POPULATION INVERSION

The Separation Maser

Of the various methods of population inversion, the simplest, in principle, is the one first used by C. H. Townes and his colleagues at Columbia University [2]. If a beam of atoms or molecules mainly in states corresponding to the two levels with energies E_2 and E_1 could be passed through some kind of filter that would pass those in the state with energy E_2 and reject those in the state with energy E_1, we should certainly obtain a condition in which the population in the upper level exceeds that in the lower, a condition we shall describe as 'emissive'. If the beam were then passed into a waveguide or resonant cavity where it could interact with radiation of appropriate frequency, amplification would be obtained by stimulated emission. This is precisely what Townes and his colleagues successfully did. In order to filter out the atoms or molecules in the ground state, some 'label' must be found to identify the states. Townes and his colleagues used ammonia molecules, which have two states separated by a small amount corresponding to a quantum of wavelength about 3 cm. The induced electric dipole moment of the molecule is different for the two states, and they may be separated by means of an inhomogeneous electric field. Such a maser is usually described as a separation maser and is illustrated in figure 4.

If sufficient amplification is available to overcome the cavity losses, the amplitude of the radiation in the cavity will build up and the system will oscillate, the amplitude building up from the random fluctuations of electric field in the cavity that are always present even when there is no input signal. It is, in fact, as an oscillator that the ammonia maser has found its most important applications. The ammonia absorption-lines are quite narrow, and the bandwidth available as an amplifier is inadequate for most purposes. When used as an oscillator, the frequency is mainly determined by the frequency of the ammonia absorption-lines (there are several due to molecular rotation) and is influenced only slightly by the tuning of the cavity. Thus the maser oscillator gives a very stable and well-defined frequency. More recently, this type of oscillator has been made to operate with atomic hydrogen at Harvard University [3], and it then provides an extremely stable frequency. It has therefore an important application as an 'atomic clock'.

The Inversion-type Maser

In order to obtain a wider bandwidth by the use of broader absorption lines, active molecules in the form of paramagnetic ions in a crystalline solid are frequently used. As previously explained, they are used in low concentration in a host crystal. If we have a single unpaired electron in the ion, the lowest state of the ion is split in a magnetic field H into two levels of energy

$$E_1 = E_0 - \mu H$$
$$\text{and } E_2 = E_0 + \mu H$$

where E_0 is the energy of the state in the absence of a magnetic field and μ is the magnetic moment of an electron. The separation $E_2 - E_1$ is therefore $2\mu H$, and for convenient values of H, the corresponding frequency ν, equal to $(E_2 - E_1)/h$, comes in the microwave region of the spectrum. Thus

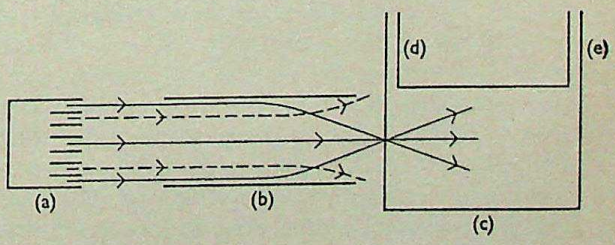

FIGURE 4 – *Separation maser.* (a) *Oven source,* (b) *separator,* (c) *resonant cavity,* (d) *microwave input,* (e) *microwave output.*

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if $H = 10^4$ oersted, $\nu = 28$ kMc/s, $\lambda \simeq 1$ cm. Now if we were to place such a crystal in a magnetic field at a low temperature in order to obtain an appreciable difference in the populations of the levels corresponding to E_1 and E_2, we should have $N_1 > N_2$.

If we could suddenly reverse the direction of the magnetic field, the levels 1 and 2 would be interchanged while their populations remained unchanged. There would then be more ions in the upper level than in the lower, and this is an emissive condition. Unfortunately, this simple method of obtaining population inversion cannot easily be put into practice, as the time taken to reverse the field is generally so long that the populations have time to relax to their equilibrium condition. It may be shown, however, that the same result can be obtained by applying a strong microwave signal of frequency a little less than that corresponding to the absorption line and sweeping it quickly, but not too quickly, to a value above that of the line. This is known as adiabatic fast-passage inversion, and microwave amplifiers have been made to operate using this technique. Such amplifiers, however, operate only in pulses, since the populations revert to their equilibrium values in a few milliseconds, and the inversion then requires regeneration. By far the most commonly used method of inversion, which has the advantage of giving continuous amplification, is that devised by N. Bloembergen of Harvard University [4]. This method uses three energy-levels.

The Three-level Maser

Let us suppose that we have three closely spaced energy-levels whose energies E_1, E_2, E_3 are such that $E_1 < E_2 < E_3$ with populations N_1, N_2, N_3. These levels are commonly derived from paramagnetic ions that have more than one unpaired electron. The Cr^{3+} ion, for example, gives four levels, three of which may be used. Ruby crystals (Cr^{3+} in Al_2O_3) are very commonly used as active material. Now suppose we apply a strong microwave signal of frequency $\nu_{31} = (E_3 - E_1)/h$ and saturate the pair of levels 1 and 3. Then we have, approximately, $N_1 = N_3$. We shall suppose the levels 2 and 3 to be closely coupled thermally, that is, they have a short relaxation time. Their populations will therefore not depart appreciably from their thermal equilibrium ratio, and we shall have $N_3 < N_2$. Since $N_1 \simeq N_3$, it follows that $N_1 < N_2$, and we obtain an emissive condition between the levels 1 and 2. Amplification at a frequency $\nu_{21} = (E_2 - E_1)/h$ may thus be obtained. This is illustrated in figure 5. The frequency ν_{31} is usually called the 'pump' frequency and ν_{21} the signal frequency; it is important to note that ν_{31} is greater than ν_{21}. If a crystal containing suitable active ions is inserted in a resonant cavity, oscillations at frequency ν_{21} may be produced if the stimulated emission exceeds the cavity losses.

This type of maser is, however, of little use as an oscillator, because its emission 'lines' are broad; its main use is as an amplifier. When attempts were being made to amplify at ever higher microwave frequencies, the fact that the signal frequency was less than the pump frequency seemed unattractive. The three-level maser amplifier turns out, however, to have an extremely low noise-level as compared with conventional microwave amplifiers, and it is in this that its great importance lies. We shall return to this point later.

The schematic arrangement for a three-level cavity-maser amplifier is shown in figure 6. The active material, usually ruby (shown shaded), is enclosed in the resonant cavity *a*. This has to be carefully designed to be resonant at both the pump frequency and signal frequency, otherwise a very high pump-power would be required. The resonance increases the field strength obtained in the active material, and is equivalent to the radiation's having many passages through the material. The signal, generally fed to the cavity along a coaxial line—a waveguide can be used—is reflected in the cavity and amplified, passing back along the same lead. The incoming and outgoing signals are separated in the 'isolator' *c*; the inputs and outputs are shown as *d* and *e*. The cavity is placed between the poles of an electromagnet *f*, in a Dewar flask *g*. In a more modern 'packaged' design, a small permanent magnet or superconducting magnet immersed in the cooling liquid with the cavity is used. Liquid helium is most commonly used for cooling, but maser amplifiers that operate, with, however, reduced efficiency, using liquid nitrogen have also been made. The pump power is fed to the cavity by a waveguide *h*. The power gain

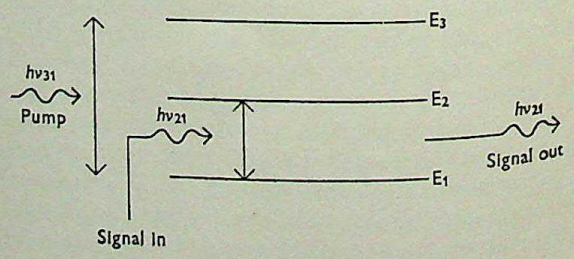

FIGURE 5 – *Scheme for three-level maser.*

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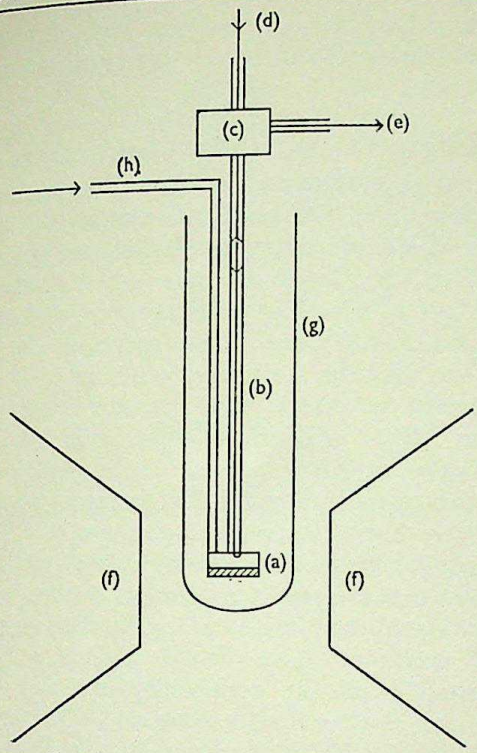

FIGURE 6 – *Arrangement of three-level cavity maser.* (a) *Cavity with active material (shown shaded)*, (b) *signal lead to cavity*, (c) *isolator*, (d) *signal input*, (e) *signal output*, (f) *magnet*, (g) *Dewar flask*, (h) *pump input.*

obtainable with an amplifier of this kind depends on the frequency bandwidth over which the amplification is effective. It can be shown that the product of the square root of the gain and the bandwidth is approximately constant. Values of approximately 50 for this constant are common, the frequency being expressed in Mc/s, and even higher figures have been obtained. For example, a power gain of 400 (26 db) may be obtained with a bandwidth of 2·5 Mc/s.

The active material, in the form of a long strip, may be enclosed in a waveguide instead of a resonant cavity. The waves travelling down the guide in one direction are amplified by interaction with the active material; for this reason the device is called a travelling-wave amplifier. In order that the waves may interact for a longer time with the active material, a special form of waveguide is used. This slows the waves down considerably in the section of the guide containing this material. An ingenious arrangement ensures that only waves travelling in one direction interact with the active material, while waves travelling in the opposite direction interact with absorbing material that is introduced into the waveguide, and so are attenuated. The pump power has also to be fed down the waveguide so as to make the amplifying material active. For this type of amplifier there is no need for an 'isolator' to separate the incoming and outcoming signal after interaction with the active material. The signal is simply fed in at one end of the waveguide and comes out amplified at the other. The isolators used with cavity amplifiers are found to produce unwanted noise, so the omission of this component is of considerable advantage [5]. The travelling-wave amplifier also proves to be very stable in operation and, as we shall see, has been used with success as a very-low-noise amplifier.

AMPLIFIER NOISE

We have not yet discussed the enormous advantage that maser amplifiers have over conventional microwave amplifiers. This is in the almost complete absence of the random fluctuations, 'noise', that set a lower limit to the signal strength that can be intelligibly received. Everyone is familiar with the background hiss in a radio receiver, or the 'grass' on a television screen, that occurs when the gain is turned up in order to try to receive a weak station. When this noise occurs, further increase of gain will not improve reception, and the limit in sensitivity is set by the signal-to-noise ratio. Some of the noise is externally generated and some produced in the amplifier itself [1]. The latter usually predominates in conventional microwave receivers. Any resistive load produces noise of a fundamental character, called Johnson noise, and a current of free electrons, being rather like a shower of rain, produces noise called shot noise. Similar noise is caused by the electrons flowing in the crystal 'detector' of a conventional microwave receiver. By avoiding this type of noise, the maser amplifier achieves a very much lower noise-value than a conventional receiver—often about 1000 times less.

NOISE TEMPERATURE

In order to specify the quality of a receiver, electronic engineers frequently use temperature as an index. The maximum noise-power in the form of Johnson noise per unit bandwidth that can be drawn from a resistive load depends only on the absolute temperature T of the load. If we say that noise corresponds to a noise temperature T_e we mean that we have the same noise power per unit bandwidth as we should get from a load at temperature T_e. T_e is therefore an index that increases linearly with noise power per unit bandwidth. For example, a conventional microwave

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receiver may have a noise temperature of 2100° K, of which only 300° K is contributed by the input load; 1800° K comes from the receiver itself, which therefore produces six times more noise power than the input. With a maser receiver, on the other hand, a noise temperature of 3° K may be obtained, so that, if used with an input load at room temperature, the receiver itself would produce only one-hundredth of the noise power of the load.

There would, of course, be no point in using so good a receiver with such a load. This would appear to indicate that maser receivers would be worth using only with cooled loads. In a sense this is so, but the load need not be artificially cooled. It turns out that, in the microwave region of the spectrum, the sky 'looks' as though it were very cold, having an effective temperature of only a few degrees K. If we have a well-designed aerial with no large back lobes that 'look' at the ground, and a low-loss feeder system, this behaves like a very cold load when the aerial is directed well above the horizon, and in this condition a maser receiver can give an enormous improvement over a conventional microwave receiver. Maser receivers have already been used with conspicuous success in this way, mainly in radio-astronomy and for communication using artificial satellites.

A PRACTICAL SYSTEM

As an example of a complete system using a maser amplifier we may consider the arrangement described by R. W. De Grasse and his colleagues [6] for use in the reception of signals from the artificial satellite in the form of a spherical balloon, known as Project Echo. The whole system achieved the remarkably low overall noise temperature of 18·5° K. This means that the noise was equivalent to what would be obtained from an ideal noiseless receiver with an input load cooled to 18·5° K. The maser amplifier had a power gain of about 3000 and operated at a wavelength of 5·3 cm. The receiver itself contributed noise corresponding to a temperature of 10·5° K. The aerial and feeder system gave 5·5° K and the effective temperature of the sky was 2·5° K. This must be regarded as quite a remarkable achievement when we remember that the best noise figure obtainable for a conventional microwave receiver at this wavelength corresponds to a noise temperature of about 900° K.

When the aerial is pointed horizontally it will receive noise radiation at about 300° K from the ground, and the advantage of the low-noise receiver will largely be lost. It is therefore for reception from elevated objects such as stars or artificial satellites that maser receivers will prove most useful.

AMPLIFICATION OF LIGHT—THE LASER

Although spontaneous emission is so much more probable than stimulated emission in the optical region of the spectrum, conditions can be found in which stimulated emission can be made to build up so as greatly to exceed spontaneous emission in intensity. If we examine the light from an electric discharge, we find that it is incoherent in phase, like random noise. The reason is that most of the light is due to spontaneous emission from atoms excited electronically, and this emission is a random process. Suppose, however, that by some means, a population inversion could be achieved for two levels whose energy difference corresponds to an optical frequency. There would then be a small probability of stimulated emission. This would indeed be small compared with the probability of spontaneous emission, since we now have $h\nu \gg kT$. On the other hand, the stimulated emission is phase-coherent, and this may give rise to a much greater intensity in spite of the small probability. This may be seen as follows. Suppose we have a large number, N, of identical radiators whose phases are randomly distributed. Then the intensity of radiation, which takes place in all directions, is simply NI, where I is the intensity from each. If, however, the radiators are coherent in phase, and the radiation is observed in a direction in which they are all in phase, the intensity will be N^2I. If N is very large, as it will be even for a gas or a solid diluted with an active impurity, a vast increase in intensity is possible. In order to achieve this, two conditions are necessary. Firstly, an emissive condition must be established, and secondly, an optical arrangement must be made such that a large number of atoms radiate in phase. We shall later discuss various methods that have been used to obtain population inversion in the optical region of the spectrum; the second condition has generally been achieved by using two parallel plates with highly reflecting surfaces, as in a Fabry-Perot interferometer. This is equivalent to the cavity used in the maser and causes the light to pass a large number of times through the active material while retaining phase coherence over a large area. One of the plates is made slightly transparent, and the phase-coherent beam emerges through this. This is illustrated in figure 7. A scheme of this kind was first proposed

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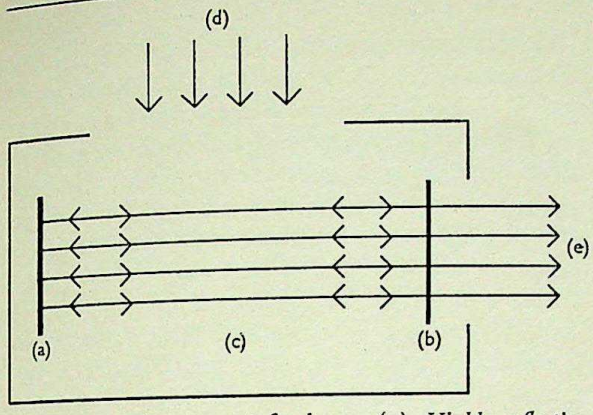

FIGURE 7 – *Arrangement for laser.* (a) *Highly reflecting opaque flat plate,* (b) *parallel highly reflecting slightly transparent flat plate,* (c) *active material between plates,* (d) *pumping radiation,* (e) *emergent phase-coherent beam.*

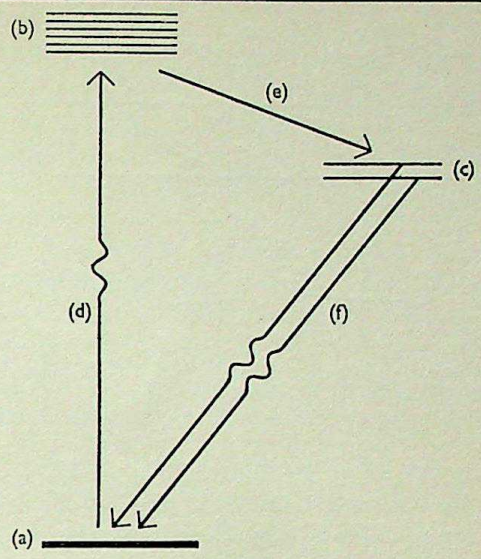

FIGURE 8 – *Level scheme for ruby laser.* (a) *Ground state,* (b) *broad band of excited states,* (c) *metastable state (doublet),* (d) *pumping transitions,* (e) *drop to metastable states,* (f) *fluorescent and stimulated emission.*

by A. L. Schawlow and C. H. Townes [7], who suggested using a vapour such as Na or K as the active material.

PULSED RUBY LASER

Stimulated emission of light was first observed by T. H. Maiman [8], using ruby as the active material, and a phase-coherent beam was obtained soon after by a group at the Bell Telephone Laboratories [9]. It is interesting that ruby should be used again as the active material in the laser as well as in the maser, though in this case naturally occurring levels of the Cr^{3+} ion are used and not the magnetically split levels used in the maser. The mechanism that is used is as follows. The pump power is used to raise chromium ions to a band of excited states that are broadened because of the interaction of the chromium ions with the atoms of the host lattice. Because this is a broad band, a lot of pumping power can be absorbed. The ions fall from this band to a metastable level, in fact a doublet, from which they return to the ground state by emitting the characteristic red fluorescence of ruby. The fluorescence consists of two narrow lines in the red region of the spectrum. This is illustrated in figure 8. If the pumping power is sufficiently high, the population of the ground state can be reduced sufficiently to make it less than that of each of the levels of the metastable state, and an emissive condition then exists. The very high pumping-power is obtained by discharging a bank of condensers through a special high-intensity flash tube. The flash lasts only for a short time, and the fluorescence that follows lasts for about one millisecond. The group in the Bell Telephone Laboratories made their interferometer for containing the radiation by grinding parallel optical flats on the ends of a ruby rod about 0·5 cm in diameter and 4 cm long. The flat ends were silvered, one heavily and one lightly, and an intense beam of red light was observed from the lightly silvered end when the intensity of the pumping flash exceeded a certain value. The arrangement is illustrated in figure 9.

Devices of this kind are currently known as lasers, by analogy with masers, the 'l' standing for light. It should, however, be pointed out that controlled amplification has not been achieved, and that the devices are coherent-light oscillators. The coherent beam from the pulsed ruby laser lasts about 0·5 msec, the total energy being about 0·2 joule. The input energy to the flash tube is about 5 kilojoules, so that the device is very inefficient as regards energy conversion. However, if the output radiation were phase-coherent over the entire end of the ruby, it could be focused to a spot whose diameter is approximately one wavelength. Unfortunately, because of irregularities in the ruby crystal, coherence is possible over only limited areas. The output beam can, however, be focused to a spot of diameter 0·1 mm. The mean output power is 0·4 kW, so that the power flux at the focused spot is about 4×10^6 W/cm^2. By focusing the sun with an $f/1$ lens, an intensity of about 500 W/cm^2 can be achieved. Thus the focused spot from the laser is about 10 000 times brighter.

Various applications of such a device can be envisaged. A pulsed ruby laser has already been

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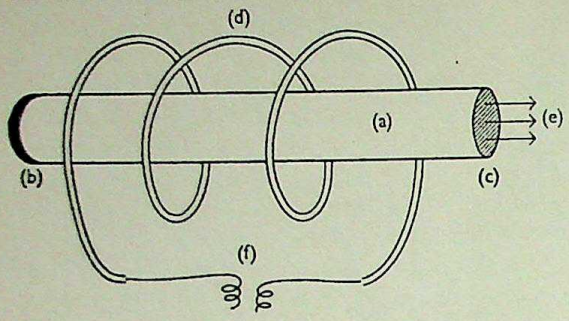

FIGURE 9 – *Pulsed ruby laser.* (a) *Ruby rod,* (b) *heavily silvered flat end,* (c) *lightly silvered flat end,* (d) *exciting flash-tube,* (e) *emergent coherent beam,* (f) *leads to condenser bank.*

used for flash micrography at the Bell Telephone Laboratories [10]. Signalling from the moon or from artificial satellites with so bright a source is also a possibility. Indeed, all sorts of intriguing possibilities suggest themselves. If longer pulses can be achieved, it should be possible to melt refractory materials. The device is in a very early stage, and the next few years will undoubtedly bring about a variety of new developments and applications.

THE COOLED LASER

A severe disadvantage of the pulsed ruby laser is the large power required for the optical pumping. This is because, in equilibrium, most of the Cr^{3+} ions are in the state to which the emissive transition is made, that is, the ground state. If an ion could be found for which a strong narrow emission line exists, with a final state lying somewhat above the ground state, the population inversion could be made more easily and with much less pumping power. The reason for this is that, if the crystal were cooled, the final state would have a very small population in equilibrium. If ΔE is the separation in energy of the final state from the ground state, the condition is that $\Delta E \gg kT$. One or two such ions are known. For example, Sm^{2+} ions in CaF_2 give an emission line in the red part of the spectrum that terminates on a level 0·045 eV above the ground state. At liquid helium temperature $\Delta E/kT = 130$, so that in equilibrium the terminal state will be virtually unpopulated. P. P. Sorokin and M. J. Stevenson [11] have used this ion to make a laser based on this principle. Stimulated emission in the near infra-red has also been obtained using U^{3+} ions in CaF_2. These lasers were also pulsed, but there is reason to believe that, because of the smaller pump power required, continuous operation might be possible.

THE GAS LASER

Continuous operation of a laser has, so far, been obtained only by using a gas mixture as the active medium. This was achieved by A. Javan and his colleagues at the Bell Telephone Laboratories [12], using a mixture of helium and neon excited by an electric discharge. The neon gas provides the active material and has a pressure of about 0·1 mm Hg; the helium gas, at a pressure of about 1 mm Hg, serves as an intermediary to provide the optical pumping. The arrangement is shown in figure 10. The gas mixture is contained in a quartz tube, *a*, with internal diameter 1·5 cm and length 80 cm, widened at the ends to accommodate two parallel Fabry-Perot plates *b*; optically flat windows, *c*, are provided at the ends for the emergent beams, *d*. The Fabry-Perot plates have to be very carefully made. They are made from silica and have highly reflecting coatings built up from no fewer than thirteen evaporated layers. They are flat to one hundredth of a wavelength at 1 μ and are placed in the gas mixture. An external micrometer adjustment enables them to be lined up so as to be accurately parallel. They reflect 98·9 per cent and transmit 0·3 per cent of the incident radiation in a narrow band of wavelengths in the near infra-red part of the spectrum. The electric discharge is excited by electrodes, *e*, carrying high-frequency currents. The exciting frequency is 28 Mc/s, and only 50 W of power is required. This is very much smaller than for the solid laser, and allows continuous operation.

The mechanism of population inversion is somewhat complex and very ingenious. Helium atoms in a gas at low pressure may readily be excited by electrons in a discharge to a metastable state lying 19·8 eV above the ground state. Because the state is metastable, a large population of atoms excited to this state can be set up in the discharge. Now the neon atom has a set of closely spaced states lying very near in energy to the metastable helium state (2S levels). The excitation energy can be transferred to these states by collisions between metastable helium atoms and neon atoms in the ground state. Collisions of this type are known as collisions of the second kind. Another closely

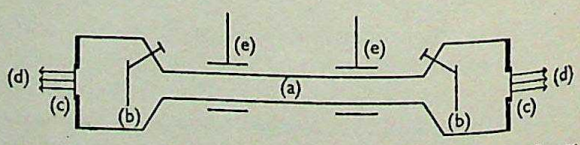

FIGURE 10 – *Arrangement of gas laser.* (a) *Gas discharge tube,* (b) *optical flats (slightly transparent),* (c) *windows,* (d) *emergent phase-coherent beams,* (e) *r.f. excitation.*

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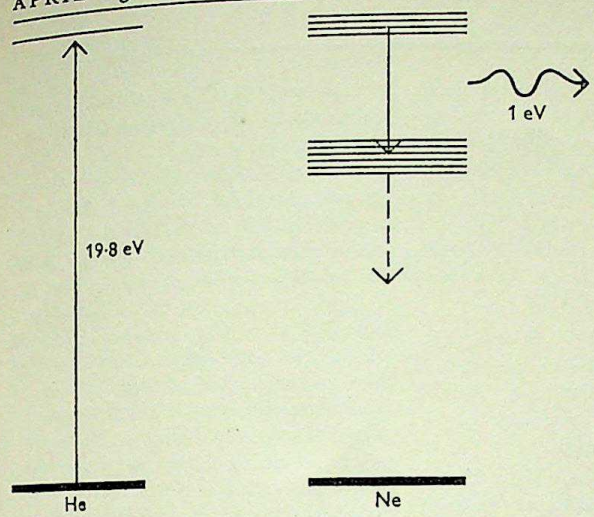

FIGURE 11 – *Level schemes for He and Ne as used in the gas laser.*

spaced set of levels (2P levels) of the neon atom lies about 1 eV below these levels, and transfer of excitation to these levels by collision is improbable because of this energy separation (see figure 11). The population of some of the 2S levels of neon may therefore under proper conditions be made to exceed that of some of the 2P levels, and this is an emissive condition. Javan and his colleagues succeeded in obtaining stimulated emission between various pairs of states associated with the 2S and 2P levels, the strongest having a wavelength of about 1·15 μ, which is in the near infra-red region of the spectrum. A coherent, nearly parallel beam emerged from the end windows, with a power of 15 mW and a diameter of 0·45 in. It was shown by interferometric measurements to be phase-coherent across most of its diameter, and its angular spread was less than one minute of arc. Another notable feature was that the spectral bandwidth of each nearly monochromatic component of the light was of the order of 80 kc/s, very much narrower than the spectral lines usually observed.

A device like the above that radiates in the infra-red is sometimes known as an 'iraser'; it is not different in principle from the laser, and we prefer to retain the latter term. There is no reason why such a device should not be made to emit in the visible region of the spectrum, as it is only a question of finding a suitable pair of levels whose population can be inverted. Nearly monochromatic sources in the infra-red are, however, almost non-existent, and the production of these may well be one of the most important applications of the laser.

Already a great deal of ingenuity has gone into devising methods for obtaining an emissive condition in a gas or vapour, and a number of other proposals have been put forward. The next few years are likely to bring forth many developments in this very exciting new field of research.

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Book reviews

DIFFERENTIAL EQUATIONS

Differential Equations, *by F. G. Tricomi (translated from the Italian by Elizabeth A. McHarg). Pp. x+273. Blackie & Son Ltd, London.* 1961. 50*s. net.*

This is the English translation of the third edition of what has become a standard and classical introduction to the modern theory of differential equations. The treatment combines clarity and rigour and uses the most modern methods that have been devised for establishing the fundamental existence theorems in this subject. The mathematical physicist will probably be most interested in the second chapter, which discusses the geometric theory of non-linear differential equations of the first order. On the other hand, although the physicist may be somewhat daunted by the detailed proofs of the theorems of the later chapters, he will probably be extremely interested in the results obtained, especially those relating to comparison methods for linear equations of the second order and to the form of asymptotic solutions. G. TEMPLE

TOPOLOGY

Topology, *by J. G. Hocking and G. S. Young. Pp. ix+374. Addison-Wesley Publishing Co. Inc., Reading, Mass., and London.* 1961. 74*s. net.*

This excellent book fills a long-felt need for a textbook suitable for a graduate course in topology. Both point-set and algebraic topology are covered, and the text is illustrated with a large number of examples. There is quite a sprinkling of exercises, and there are many diagrams and an extensive bibliography. It is assumed that the student has been introduced to set theory, group theory, and the theory of functions.

The first chapter deals with topological spaces and functions, and then two chapters are given to the development of point-set topology. The fourth chapter introduces homotopy theory. In the fifth chapter the study of algebraic topology commences with an account of polytopes and triangulated spaces, and there then is a chapter on simplicial homology theory. Relative homology and cohomology are studied in chapter 7, and the book closes with a rather advanced chapter on general homology theories.

Not every student of pure mathematics finds topology an easy subject, and this may be partly because few accounts are available at a reasonable level. The subject is undoubtedly sophisticated compared with some of the more classical branches of mathematics, and for a long time we have lacked a text that starts at the proper level. This book should go a long way towards repairing this omission. L. S. GODDARD

ASTROPHYSICS

Basic Physics of the Solar System, *by V. M. Blanco and S. W. McCuskey. Pp. xi+307. Addison-Wesley Publishing Co. Inc., Reading, Mass., and London.* 1961. 64*s. net.*

The main part of this book may be accepted as a supplementary textbook on the fundamental aspects of the solar system. It is assumed that the reader has the knowledge of physics and mathematics that might be expected of an engineer or non-specialized astronomer. This assumption has enabled the authors to go considerably further in their theoretical discussions than in a descriptive book. Orbital theory is taken up to the stage of deriving planetary observations from orbital elements, but the reverse problem of determining elements is discussed only in simplified cases. The reader is introduced to the theory of the three-body and n-body problems, and to the perturbations of artificial satellites by terrestrial oblateness and atmospheric resistance.

The more astrophysical parts of the book touch on so many diverse matters in a short space that the treatment is necessarily sketchy. It aims at giving the reader some stimulating quantitative ideas rather than a descriptive coverage. C. W. ALLEN

QUANTUM MECHANICS

Quantum Mechanics, *Vol. I, by A. Messiah (translated by G. M. Temmer). Pp. xv+504. North-Holland Publishing Co., Amsterdam.* 1961. £5 *net.*

There can be little doubt that the two volumes of Messiah's 'Quantum Mechanics' comprise the most thorough and systematic exposition of quantum theory, up to an introduction to quantization of the electromagnetic field, yet written or, perhaps, ever likely to be. The book, based on the author's course at the *Centre d'Etudes Nucléaires* at Saclay, was published in French some three years ago, and the North-Holland Publishing Company is to be congratulated on having produced this English translation. So far we have the first volume only, but the second is promised in due course.

Although technical details are not over-emphasized, the book does presuppose a more mature attitude to mathematics than is commonly found in English physics students. For this reason, it is perhaps not to be recommended as a first introduction to its field. But as an exposition not only of the formalism but also of the ideas of quantum theory it is unsurpassed. Its emphasis on the correspondence principle is particularly welcome. And there are excellent problems at the end of each chapter. G. S. RUSHBROOKE

PLASMA PHYSICS

Plasmas and Controlled Fusion, *by D. J. Rose and M. Clark, Jr. Pp. xiv+493. The M.I.T. Press, Massachussets Institute of Technology; John Wiley & Sons, New York and London.* 1961. 86*s. net.*

This book is written as a textbook for graduate study, and although it starts with first principles, it is not an elementary book. It is recommended for post-graduate teaching in universities and as a reference book for the many scientists and engineers now becoming involved in this expanding research field.

The first twelve chapters provide a course in plasma physics, hydromagnetics, and gaseous electronics, and the choice of subject matter, presentation, and development reflect the experience of the authors in teaching the subject to graduates. Each chapter ends with a list of problems and with references for further study. The remaining chapters, in which the fundamental concepts are applied to some of the current fusion experiments, are necessarily selective and, since none of the present approaches is yet successful, incomplete. The inclusion of these last chapters can also be justified as a means of encouraging graduates to enter this field of research. However, whereas the first half of the book has a permanent value, the second half will ultimately become out of date, and, since space in the book does not permit a full record, even its historical value will not be great. The authors are well aware of this problem and have tried to use the present struggles to illustrate the fundamental concepts.

The whole book is a valuable contribution to the literature of plasmas and controlled fusion research.

J. B. ADAMS

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INTERMOLECULAR FORCES

States of Matter, *by E. A. Moelwyn-Hughes. Pp. viii+100. Oliver and Boyd, Edinburgh.* 1961. 15*s. net.*

This a very enjoyable and unusual little book. The states of matter considered are seven, that is the crystalline, the gaseous, the metallic, the liquid, the dissolved, the ionic, and the interfacial, and a unified treatment is given on the basis of the concept of intermolecular forces. The theory of such forces is developed in as simple a way as possible in the first chapter and then applied almost directly to give an account of current ideas on the various states of matter. Considerable information is provided on selected topics. This is thus one of the lamentably few attempts to bring together, under a unifying point of view, subjects generally treated in different branches of physics and chemistry. As such it is very welcome, and while the specialist in any of the fields covered may perhaps in places consider the treatment as superficial, one-sided, or incomplete, non-specialists will read it with profit, and students will find it a very useful complement to standard lecture courses.

L. PINCHERLE

PHYSICAL CHEMISTRY

Physical Chemistry (*second revised edition*), *by E. A. Moelwyn-Hughes. Pp. vii+1333, Pergamon Press Ltd, Oxford.* 1961. 84*s. net.*

This interesting textbook has retained its clear format and attractive presentation throughout the present edition. The opportunity has been taken to make a number of corrections and additions and some deletions, with the overall result that the text has increased in length by about 35 pages. The main additions are found in the chapters on intermolecular energies, liquid state, dissolved state, interfacial state, and kinetics of reactions in gaseous systems. An additional short appendix has been added to the previous ten mathematical or numerical appendices. One may perhaps comment on the use of the word 'state' in such phrases as 'the interfacial state' or 'the dissolved state'. No fewer than seven of these 'states' are referred to in place of the usual three physical states of terrestrial matter—liquid, solid, and gaseous. This seems an unnecessary elaboration of the term, especially as chapter headings. It is somewhat confusing when used in so many connections. One is glad to see deletion of certain statements, such as that regarding the impossibility of measuring the wavelength associated with protons on page 137 of the old text, or that regarding the degree of dimerization of iodine in certain solutions on page 737. An error on page 158 of the new text (eq. 83) is not yet corrected.

Such criticisms as these, however, are small ones, and Dr Moelwyn-Hughes has improved this valuable and readable text, first published in 1957.

R. M. BARRER

INTERFACIAL PHENOMENA

Interfacial Phenomena, *by J. T. Davies and Sir Eric K. Rideal. Pp. xiii+474. Academic Press, New York and London.* 1961. £5 *net.*

The study of substances in the two-dimensional state at surfaces and interfaces has long been recognized as an important branch of physical chemistry. The subject is not only of fundamental interest but has great relevance to a variety of processes employed in industry and to an understanding of biological reactions where interfaces are involved. This volume gives an account that is both comprehensive and authoritative and that contains a judicious blending of the older work with more recent developments. The chapters about electrostatic and electrokinetic phenomena are particularly valuable, although the treatment will not be easy to follow for readers who lack a good basic knowledge of physical chemistry. The book should prove most stimulating to those who are concerned with either the academic or the industrial aspects of surface chemistry.

C. KEMBALL

SPECTROCHEMICAL ANALYSIS

Spectrochemical Analysis, *second revised edition, by L. H. Ahrens and S. R. Taylor. Pp. xxiii+454. Pergamon Press Ltd, Oxford; Addison-Wesley Publishing Co. Inc., Reading, Mass.* 1961. £5 5*s. net.*

In the first edition, Professor Ahrens wisely chose to cover a selected field in which he had very wide experience. The worn and well-thumbed state of the copies found in so many spectrographic laboratories is sufficient witness to its success as a practical guide to the spectrographic determination of elements in geological and related materials.

The second edition, revised with the aid of Dr S. R. Taylor, happily shows no basic changes. Beyond comprehensively bringing the text up to date and rearranging some of the material, new chapters have been added on enrichment methods and operating procedures. The sections on semi-quantitative analysis and the accurate determination of major elements have been much expanded to include the important progress made in the last ten years.

These revisions and additions have further increased its already great practical value. Even the minor improvements, such as ending the petty annoyance of having to convert trace concentrations from percentages to parts per million, and the inclusion of more energy-level data, will be much appreciated by the laboratory worker. Whether he be geochemist or spectrographer, this edition will be an essential part of his reference library.

K. G. CARR-BRION

OCEANOGRAPHY

Oceanography. Invited Lectures presented at the International Oceanographic Congress, New York, 1959. *Edited by Mary Sears. Pp. xi+654. American Association for the Advancement of Science, Washington, D.C.; Bailey Bros. and Swinfen Ltd, London.* 1961. £7 10*s. net.*

The programme of the first International Oceanographic Congress held in the United Nations Headquarters, New York, from 31st August to 11th September 1959, was arranged round five main themes: history of the oceans; populations of the sea; boundaries of the sea; and cycles of organic and inorganic substances. Two morning sessions were devoted to each topic, and invited speakers were asked to review contemporary progress. Dr Mary Sears, the chief organizer of the Congress, has edited these talks in this attractive volume.

It is a tremendous record of work and mobilization of ideas, more than up to date, with plenty of well-judged prediction, although every now and then an expert conscious of the difficulty and achievements of his own subject indulges in forgivable speculation about impinging topics that he does not understand so well. It is difficult to pick out outstanding examples from the thirty contributions, but those of Arrhenius, Bullard, Eriksson, Ewing, Malkus, Thorson, and Welander call for special mention.

G. E. R. DEACON

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PLANT HORMONES

Les phytohormones de croissance, *by P.-E. Pilet. Pp.* 774. *Masson et Cie, Paris.* 1961. *Paper covers* 100 *NF; cloth,* 110 *NF.*

It needs some courage to write a book on plant hormones these days—Professor Pilet estimates that in the last thirty years some 26 000 relevant papers have been published! He has approached this almost superhuman task rather differently from the way others have done recently, and has produced a useful, individual, and interesting book.

Let me first list some of the good features of the book. It covers all aspects of the chemistry, biochemistry, and physiology of the natural indole-auxins and their synthetic analogues in great detail. The chapters on methods of bioassay are particularly valuable. The book is lavishly supplied with attractive text figures and useful tabular matter. It has 610 pages of succinct text, over 100 pages of bibliography, and a good index. It is a first-rate source of references to original papers.

But it has weak features too; I will mention one. The gibberellins and cell-division hormones (kinetin) are dismissed in fourteen pages. The whole of recent work on growth regulation in plants supports the view that auxins, gibberellins, and cell-division hormones, and perhaps others yet to be discovered, are of equal status, and that plant-growth is regulated by their interaction rather than by single hormones. Professor Pilet, by his almost exclusive concentration on the auxins, gives a rather old-fashioned and unbalanced view.

Thus, while I feel that this is not as good a book as it could have been, it is one that most plant physiologists could consult with advantage, not least because it deals with recent work on the continent of Europe far more adequately than English and American text-books have done. P. W. BRIAN

FUNGI

British Cup Fungi and their Allies, *by R. W. G. Dennis. Pp. xxiv*+280. *The Ray Society, London.* 1960. 80*s. net.*

'British Ascomycetes' would have been a more accurate title, as much more space is given to the different orders than to those Discomycetes—Pezizales and larger Helotiales—sometimes called Cup Fungi because of the shape of the fruit body.

After a short introduction, the book plunges at once into a purely systematic treatment. The descriptions are clear and concise and, on the whole, satisfactory. Many recently described genera and species are included and thus made readily available for those unable to consult original papers.

A special feature is the large number of plates. Twenty are black-and-white with many figures, mostly of the text-book type and sufficient for their purpose. The forty coloured plates leave much to be desired, both in draughtmanship and colour: more care has been given to their reproduction than to their preparation.

The author's intention was to write a semi-popular illustrated book, but there is no glossary and little to help a beginner. Moreover, the nomenclature adopted, in the attempt at modernity, is bewildering, for new names are proposed; this ensures that mycologists will need to consult the work. It will serve as a useful handbook for them.

J. RAMSBOTTOM

EUCALYPTS

Eucalypts, *by A. R. Penfold and J. L. Willis. Pp. xx*+551. *Leonard Hill (Books) Ltd, London; Interscience Publishers Inc., New York.* 1961. 80*s. net.*

This is a very interesting and informative book. It is not primarily intended for the professional botanist, but does bring together a wealth of information from the widely scattered, often rather inaccessible literature on this subject. The large genus *Eucalyptus* (over 600 species) is almost entirely Australian in its distribution, but is now widely grown in warm temperate to tropical countries. The authors, both known for their work on the chemistry of eucalypt products, have brought together in one volume information on the botany, chemistry, cultivation, and utilization of these trees. Wisely they have adopted the current classification in W. F. Blakeley's 'Key', which they reprint, and have thus avoided vexed questions of taxonomy of this difficult genus. The longest chapter gives 'descriptions and digests' of the 633 species and varieties listed by Blakeley, 150 of which are redescribed on a uniform system. This offers an easier comparison of the different species than does reference to the original diagnosis, for which, however, no citation is given. There are five distribution maps. Chapters are devoted to such diverse topics as honey flora, essential oils, and cultivation and miscellaneous uses, including paper and pulp, fibreboard, wood distillation, and firewood.

In 1953 F.A.O., with the co-operation of the Commonwealth Forest Resources Division, organized a eucalyptus study tour in Australia. Twenty-six different countries were represented. The final report of the World Eucalyptus Conference (Rome, 1956), supplemented by observations made by the senior author in South America and China, makes the chapter on eucalypts as exotics particularly valuable.

There is a good bibliography and a very detailed index. The absence of any cross references in the text is a drawback, and makes frequent reference to the index essential when reading. There are 61 good half-tone plates, 28 of which are copied from the eight quarto volumes of Maiden's critical revision. T. G. B. OSBORN

HISTORY OF CHEMISTRY

A History of Chemistry, *Vol. II, by J. R. Partington. Pp. xxiv*+795. *Macmillan & Co. Ltd, London; St Martin's Press, New York.* 1961. £5 5*s. net.*

This is the second volume, and the first to appear, of a four-volume history of chemistry that has long been in preparation by Professor J. R. Partington. Volumes II and III, the latter already in the press, cover the period from about 1500 to about 1800, and there is therefore some inevitable overlap into the eighteenth century in this second volume. The work is planned as an enlargement, and a very considerable enlargement, of the author's well-known 'Short history of chemistry'. The contents of the present volume are mostly set out as a series of studies in the lives and works of the chemists whose labours fall within the respective periods dealt with, much on the same lines, as in the late Professor George Sarton's 'Introduction to the history of science', a work which, as its distinguished author always insisted, was not a history, but an introduction, as its title stated. Here also we have a bio-bibliographical or biblio-biographical encyclopaedic compilation, stored with references exhaustively gathered with Professor Partington's characteristic and unrivalled industry, a great treasure-house of historical data that will long prove invaluable to historians of chemistry. DOUGLAS McKIE

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Some books received

(*Note.* Mention of a book on this page does not preclude subsequent review.)

GENERAL SCIENCE

Œuvres scientifiques complètes, *by Frédéric and Irène Joliot-Curie. Pp. viii+915. Presses Universitaires de France.* 1961. 100 *NF.*

MATHEMATICS

Elementary Differential Equations, *by W. T. Martin and E. Reissner (second edition). Pp. xiii+331. Addison-Wesley Publishing Company Inc., Reading, Mass. and London.* 1961. 51*s. net.*

Mathematical Programming, *by S. Vajda. Pp. ix + 310. Addison-Wesley Publishing Co. Inc., Reading, Mass. and London.* 1961. 68*s. net.*

ASTRONOMY

Astronomical Photography, from the Daguerreotype to the Electron Camera, by *Gérard de Vaucouleurs (translated by R. Wright). Pp.* 94. Faber & Faber, London. 1961. 25*s.*

PHYSICS

Absorption Spectra in the Ultraviolet and Visible Region, *by L. Lang (second revised edition).* Introduction, *pp.* 80; *Vol.* I, *pp.* 414, $18.00 (*with introduction*); *Vol.* II, *pp.* 408 $16.00. *Hungarian Academy of Sciences, Budapest.* 1961.

Thermoelectricity: Science and Engineering, *by Robert R. Heikes and Roland W. Ure, Jr. Pp. xi+576. Interscience Publishers Inc., New York.* 1961. 139*s.*

CHEMISTRY

Chimie physique, *by Guy Emschwiller. Vol. I,* Thermodyamique chimique. Equilibres gazeux, *pp. xix+438,* 1959. 24 *NF. Vol. II.* Équilibres en solutions — Phénomènes de surface (*second edition*), *pp. viii+441–946.* 1961. 32 *NF. Vol. III.* Cinétique chimique — structure des molécules (*second edition*), *pp. xv+949–1623.* 1961. 50 *NF.*

A Guidebook to Mechanism in Organic Chemistry, *by P. Sykes. Pp. xiii+247. Longmans, Green & Co. Ltd, London.* 1961. 21*s. net.*

Organic Peroxides—their Formation and Reactions, *by E. G. E. Hawkins. Pp. xiv+434. E. & F. N. Spon Ltd, London.* 1961. 80*s. net.*

Organic Peroxides, *by A. G. Davies. Pp. x+215. Butterworth & Co. Ltd, London.* 1961. 50*s. net.*

Physical Chemistry, *by F. Daniels and Robert A. Alberty (second edition). Pp. x+744. John Wiley & Sons, London and New York.* 1961. 70*s. net.*

Quantitative Organic Microanalysis, *by Al Steyermark (second edition, revised and enlarged). Pp. xvii+665. Academic Press Inc., New York.* 1961. 118*s.*

Reports on the Progress of Applied Chemistry, *Vol. XLV.* 1960. *Pp.* 775. *Society of Chemical Industry, London.*

Rules for I.U.P.A.C. Notation for Organic Compounds, *issued by the Commission on Codification, Ciphering and Punched Card Techniques. Pp. ix+107. Longmans, Green & Co. Ltd, London.* 1961. 25*s. net.*

OCEANOGRAPHY

Océanographie biologique et biologie marine, *Vol. I, by J. M. Pérès. Pp. viii+541. Presses Universitaires de France, Paris.* 1961. 30 *NF.*

BIOLOGY

Anthropology in India, *by L. A. Krishna Iyer and L. K. Bala Ratnam. Pp. xi+257. Bharatiya Vidya Bhavan, Chaupatty, Bombay.* 1961. 17*s.* 6*d. net.*

The Chromosomes, *by M. J. D. White. Pp. viii+188. Methuen & Co. Ltd, London.* 1961. 15*s. net.*

Industrial Microbiology, *by A. H. Rose. Pp. viii+286. Butterworth & Co. Ltd, London.* 1961. 60*s. net.*

Introduction to Soil Microbiology, *by M. Alexander. Pp. x+472. John Wiley & Sons Ltd, London and New York.* 1961. 76*s.*

Synthesis of Molecular and Cellular Structure, *edited by Dorothea Rudnick. Pp. viii+252. The Ronald Press Company, New York.* 1961. $9.

Tools of Biological Research, *Third Series, edited by H. J. B. Atkins. Pp. xii+155. Blackwell Scientific Publications.* 1961. 37*s.* 6*d. net.*

BOTANY

Flora of New Zealand, *Vol. I, by H. H. Allan. Pp. liv+1085. R. E. Owen, Government Printer, Wellington, New Zealand.* 1961. £5 5*s. net.*

ZOOLOGY

Bird Study, *by Andrew J. Berger. Pp. xi+389. John Wiley & Sons, London and New York.* 1961. 72*s. net.*

Cambridge Monographs in Experimental Biology, *No.* 12: Bird Song, *by W. H. Thorpe. Pp. xi+143. Cambridge University Press, London.* 1961. 20*s. net.*

Land Invertebrates, *by J. L. Cloudsley-Thompson and J. Sankey. Pp.* 156. *Methuen & Co. Ltd, London.* 1961. 16*s. net.*

Die Landmilben der Schweiz, *by J. Schweizer, Vol. LXXXIV. Pp. vii+207. Schweizerische Naturforschende Gesellschaft. Kommissionsverlag Gebr. Fretz AG, Zürich.* 1961.

MEDICINE

A Ciba Foundation Symposium on the Nature of Sleep, *edited by G. E. W. Wolstenholme and Maeve O'Connor. Pp. xii+416. J. & A. Churchill Ltd, London.* 1961. 50*s. net.*

Problems of the Physiology of the Processes of Fatigue and Recovery, *edited by the Academy of Sciences of the Ukrainian S.S.R. (translated from the Russian). Pp.* 264. *The Israel Program for Scientific Translations, Jerusalem.* 1960. 60*s. net.*

Problems of Pulmonary Circulation, Ciba Foundation Study Group No. 8, *edited by A.V.S. de Reuck and Maeve O'Connor. Pp. viii + 96. J. & A. Churchill Ltd, London.* 1961. 12*s.* 6*d. net.*

METALLURGY

Extractive Metallurgy of Copper, Nickel, and Cobalt, *edited by Paul Queneau. Pp. xv+647. Interscience Publishers Inc., New York and London.* 1961. $22.50.

Metallurgical Society Conference Series, *Vol.* 10, Columbium Metallurgy, *edited by D. L. Douglass and F. W. Kunz. Pp. xvi+746. Interscience Publishers Inc., New York and London.* 1961. $26. £9 16*s. net.*

TECHNOLOGY

The Chemistry and Technology of Tobacco, *edited by N. I. Gavrilov. Vol. III (translated from the Russian). Pp.* 768. *Program for Scientific Translations, Israel, Jerusalem.* 1961. £5. 5*s. net.*

Control of Airborne Dust, *by W. D. Bamford. Pp. viii+479. The British Cast Iron Research Association, Birmingham.* 1961. 84*s. net.*

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Fifty years of X-ray crystal analysis

When, in 1895, Röntgen discovered the new rays that penetrated solid bodies opaque to light, it at once occurred to him that they might be a novel kind of light, and he carried out a variety of experiments to test whether they were indeed of this nature. The argument at the basis of one of his tests was that a substance which in continuous form is transparent will, in powder form, allow little light to pass, on account of the refraction and reflection effected by each particle. He therefore compared the passage of his X-rays through a slab of rocksalt and through an equal mass of the same substance finely powdered, and found no difference of behaviour. No doubt he worked with the normal wide beam from his tube; if only he had used a narrow beam, he might, acute observer as he was, even with his weak tube have discovered the diffraction of the rays by the crystal and by the powder method.

The discovery, of course, created a sensation and the nature of the rays was discussed at great length. George Gabriel Stokes, then seventy-six years old, in a letter written a few months after the discovery, correctly conjectured that they were 'transverse vibrations of extreme frequency'. A little later he put forward the theory that they were a narrow pulse propagated in the ether as a result of the sudden stoppage of the electron, and J. J. Thomson took up and developed this concept. Barkla, by considering the scattering of rays themselves scattered from the primary beam, showed that X-rays had polarization properties. Haga and Wind, to test the wave nature of the rays, tried to diffract them by passing them through an extremely narrow wedge-shaped slit, and believed that they had found wave effects close to the end. Others had carried out similar diffraction experiments. These were naturally much discussed, the general conclusion being that, if waves were in question, their length must be of the order of 10^{-8} or 10^{-9} cm.

On the other hand the rays had certain particle properties, as demonstrated in particular by W. H. Bragg, who believed them, on good evidence, to consist of doublets of positive and negative electricity. The implications of the quantum theory of radiation, which requires the coexistence of both particle and wave properties, were not recognized in those days.

In 1912, then, as a result of sixteen years of intensive work there was much debate, but no certain conclusions, as to the nature of X-rays. In that year it occurred to Max von Laue, who died in 1960, that the conjectured wavelength of X-rays was shorter than the distance between the regularly spaced atoms in crystals, and that therefore their passage through crystals should produce diffraction effects. As is well known, he suggested to Friedrich and Knipping that they should attempt to establish such effects experimentally. With appropriate crystals and disposition they finally obtained the beautiful symmetrical patterns of diffraction spots that are justly celebrated. These Laue, expert in optical theory, explained as diffraction effects produced on short waves by a three-dimensional grating. This was convincing proof of the wave nature of X-rays and of their interaction with crystal structures.

It is noteworthy that what first directed Laue's attention to crystals was a question by P. P. Ewald, then a young man of twenty-four, concerning a problem in crystal optics on which he was working. Ewald was later responsible for outstanding advances in the subject of X-ray crystallography and was chief organizer and President of the Commemoration Meeting held by the International Union of Crystallography this summer at Munich to celebrate fifty years of X-ray diffraction.

Laue had established that crystals could demonstrate the nature of X-rays, but it does not seem to have occurred to him that X-rays could be used to investigate the structure of crystals. W. H. Bragg, then professor at Leeds, had carried out much important work on the ionization produced by characteristic X-rays and was an expert at handling both the not very tractable X-ray tubes of the period and the ionization chamber. His son, W. L. Bragg, twenty-two years old at the time of the Laue discovery, happened to be interested in crystal structure, an unusual concern for a young physicist in those days. The younger Bragg conceived the fundamental notion of the reflection of X-rays of a given wavelength from parallel crystal planes rich in atoms. His father developed the X-ray spectrometer, with which the intensity of the reflected rays was measured by an ionization chamber. In the years 1913 and 1914, working together and separately, father and son laid the foundations of the study of crystal structure by X-rays. W. L. Bragg worked out the crystal structure of simple examples such as sodium chloride, zinc blende, and iron pyrites; together they solved the then difficult problem of the structure of diamond. The way was shown.

Other great discoveries came in those two years.

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C. G. Darwin showed the important part played by imperfections of crystal structure, a generality which today finds expression in the dislocations that are so much invoked in connection with the mechanical properties of metals. H. G. Moseley examined systematically by the Bragg method with photographic recording the X-ray spectra of the elements and established the fundamental importance of the atomic number. Bohr put forward his epoch-making quantum theory of spectra. Then came the first world war.

After the war, the study of crystal structure developed with great rapidity, with many new techniques. For instance, the method of using a finely powdered crystal, worked out by Debye and Scherrer during the war and developed independently by Hull, found important application, and later came the method of reflecting the rays back from the surface, extensively used for investigating metals. Until recent times the photographic method, which can be made an effective record of intensity, was almost exclusively used. The electron density on the different planes was measured from reflection records and from it a Fourier synthesis constructed: elaborate development of Fourier methods, first adumbrated by W. H. Bragg, is a feature of some of the most striking modern work. While the older methods have a certain resemblance to the jig-saw puzzle method of making a map, the new methods may be likened to a trigonometrical survey.

The research school of W. L. Bragg at Manchester after the first world war was occupied with the structure of inorganic crystals of increasing complexity, exemplified by the silicates, and here Bradley achieved great things with metals and alloys, on which pioneering work was also carried out in Sweden. Meanwhile W. H. Bragg's school at University College, London, led the way in the investigation of the structure of organic crystals of familiar type, such as anthracene. This organic work was afterwards continued by W. H. Bragg and his school at the Royal Institution, where such figures as Kathleen Lonsdale, Desmond Bernal, and William Astbury developed their powers. The structure of long-chain compounds and fatty acids was, for instance, investigated.

This fruitful attack on the structural problems of organic chemistry was, in a way, a natural sequel to the investigation of inorganic crystals. Less to be anticipated, perhaps, was the success of the method with products of the living body, such as hair, horn, and wool. Astbury's work at Leeds was occupied with the structure of such substances, particularly wool, under varied conditions of humidity and temperature, problems which he solved in terms of the folding and unfolding of polypeptide chains. This was a beginning of the application of the method to fundamental biological questions. Brief reference has already been made to the X-ray study of metals, which has had most important results both for science and industry.

The most astonishing development of the X-ray method since the Second World War has been the attack on the structure of some of the giant molecules with which the biologist is concerned. The determination of the structure of vitamin B_{12}, which is an asymmetric molecule of six elements and 177 atoms, by Dorothy Hodgkin was a task of extraordinary difficulty, the success of which was mainly due to X-ray analysis. Even more astonishing, perhaps, is what has been achieved by Perutz and Kendrew, applying the latest methods to such substances as the proteins, the number of atoms in which is of the order of 10 000. Haemoglobin and deoxyribonucleic acid have been structurally analysed by Crick and Watson and much has been done on the structure of muscle. The methods used involve a narrow incident beam and ionization measurement of the diffraction, so that we can say, in a sense, that the old X-ray spectrometer has come to its own again, but in three dimensions, and with such adjuncts as extraordinary sensitiveness and automatic adjustment and registration.

It is a long journey from the table full of apparatus and the slide rule that were concerned in the pioneering discoveries to the stage represented by formidable modern X-ray installations; by such recent books as *Computing Methods and the Phase Problem in X-ray Crystal Analysis;* and by periodicals such as *Acta Cristallographica.* Today's towering models of protein molecules seem to the little sphere-and-rod inorganic models of forty years ago as the thirty-storey hotel and office blocks of today, with their carefully dispersed units held together by subtleties of structure, to small but well-built suburban houses which likewise comprise doors, windows, and a roof. The clicking of the calculating machine has replaced the soft sound of the scribbling pen, the interminable lengths of punched paper the table of logarithms. But the spirit of discovery that animates the young research workers of today, engaged, for instance, on some detail of these highly complex proteins, is no less active than, and is of the same kind as, that which drove their forerunners to reveal the structures of rocksalt and anthracene.

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Explosion in liquids and solids

F. P. BOWDEN and A. D. YOFFE

The decomposition of an explosive crystal presents problems of considerable theoretical interest, as well as being of great practical importance. This decomposition may begin slowly and then suddenly pass over to a high-speed detonation that is complete in a millionth of a second. The experimental investigation of these reactions involving high temperature and pressure is difficult. The early stages have been successfully studied; this article discusses the work on explosive compounds and their decomposition and detonation.

An explosion can be defined as any fast chemical reaction forming gas at a high pressure, and a detonation as a reaction propagated at a speed greater than the velocity of sound in the medium in which the reaction takes place; it will, therefore, be associated with a shock wave. The conditions needed to produce an explosion or a detonation in liquids and solids have been extensively studied in recent years—the stimulus has come from the need to develop new rocket fuels, from the use of explosives in nuclear devices, and from the extensive use of explosives in many scientific fields as sources of high pressure and temperature and for the production of intense shock waves. Since detonation involves extreme conditions of temperature and pressure, the experimental difficulties in studying these reactions are considerable. Furthermore, the times available for recording the transient events are short, less than a microsecond, the thickness of the zone in which the reaction takes place may be less than a millimetre, and the hot gaseous products are regions where the density of electrons and ions is high. It is therefore not surprising that we know very little about the detonation process, and that the experimental results are difficult to interpret even in terms of concepts that have been applied successfully in other branches of solid-state physics and chemistry.

It has, however, proved possible to study in some detail the early stages of the explosion process [1]. This article discusses experiments that have been carried out, mainly in the laboratory for the Physics and Chemistry of Solids at the Cavendish Laboratory, Cambridge, on the initiation and growth of explosion in liquids and solids, and on the initial stages of detonation.

One important study in this field is that of the stability of solids, and it will be useful to consider briefly the different approaches that are being made to this problem.

F. P. BOWDEN
C.B.E., Sc.D., F.R.S.

Was born in Tasmania and graduated in physics there before coming to Cambridge in 1927 as an 1851 Exhibitioner. He is now Professorial Fellow of Gonville and Caius College, and Reader and Director of the Laboratory for the Physics and Chemistry of Solids, which is part of the Cavendish Laboratory. His researches deal with the strength and surface properties of solids and with decomposition in the solid state.

A. D. YOFFE
Sc.D.

Studied at the University of Melbourne and Trinity College, Cambridge. During the war he worked in the Commonwealth Scientific Industrial Research Organisation at Melbourne, and from 1951 to 1954 carried out research in the Isotope Department, Weizmann Institute of Science. At present he is Assistant Director of Research in the Laboratory for the Physics and Chemistry of Solids. His main interests in research have been liquid surfaces, explosion in gases, liquids, and solids, and the relation between structure and stability of solids.

THE CONNECTION BETWEEN THE STABILITY OF SOLIDS AND THEIR STRUCTURE

The simplest solids from this point of view are the azides, acetylides, and oxalates [1, 2], as these decompose to give a small number of products. If we consider a series of ionic azides such as KN_3, TlN_3, AgN_3, and CuN_3, we have to explain why potassium azide is relatively stable to heat, light, and shock, whereas silver azide and cuprous azide readily explode and detonate. In this series, the stability decreases as the ionization potential of the metal increases. An examination of the structure of those azides where we are not concerned with *d*-orbitals shows that there are deviations from the ionic type of lattice as the ionization potential of the metal increases [2]. At the same time, a number of optical and electrical properties of the solids change. Measurements of the absorption spectra show that the position of the optical-absorption edge shifts to longer

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wave-lengths along the series KN_3, TlN_3, AgN_3, CuN_3. The refractive indices also increase. For example, the refractive index of KN_3 is about 1·5 for wave-lengths well removed from the optical-absorption edge, while the corresponding value for TlN_3 is about 1·8–1·9 and for AgN_3 it is about 2·2. Furthermore, whereas photoconductivity is a complicated process with pure KN_3 [3], both TlN_3 and AgN_3 are good photoconductors when irradiated with light close to the optical-absorption edge. The significance of these variations becomes more clear in terms of the theory of solid-state physics. Electrons of the atoms of a solid do not have the same energies as those of an individual free atom. They interact, so that the individual sharp energy-levels are broadened into a range of levels, called a band. This process will, of course, be most marked for the outer, valence, electrons. In semiconductors and insulators the valence band is separated by a gap from the conduction band, where the electrons are available for carrying a current. Electrons must be excited by heat or light if they are to cross the gap.

The measurements of optical properties, and, for example, of low-frequency dielectric constants and electrical conductivities, have made it possible to estimate the electron energy-levels in the solid. It has been shown that the energy gap between the valence band and the conduction band decreases along the series under consideration. The same results may also be obtained from theoretical considerations. The band gap in ionic solids is related to the activation energies obtained from experiments on thermal and photochemical decomposition.

We can therefore explain in a fairly simple way the decrease in stability along the series. From these results we can also outline a mechanism for the slow decomposition of the azides by both heat and light.

The overall equation for the decomposition can be represented by

$$2MN_3 \rightarrow 2M + 3N_2$$

Decomposition by heat develops according to the following overall scheme, involving an electron-transfer mechanism. The first stage is the formation of a positive hole, that is, an electron deficiency in the valence band, and the transfer of an electron to the conduction band. When a positive hole is localized on an azide ion, it becomes an azide radical, and the next stage in the decomposition is the reaction of two positive holes at the surface to form nitrogen; the final reaction is the absorption of a positive ion at a metal nucleus, and electron capture:

$$N_3^- = N_3 + e \qquad \ldots (1)$$

$$2N_3 \rightarrow 3N_2 \qquad \ldots (2)$$

$$M_n + M^+ + e \rightarrow M_{n+1} \qquad \ldots (3)$$

Several paths are open for decomposition by light. One involves the absorption by the crystal of a sufficiently energetic photon to raise an electron from the filled band, the valence band, to the conduction band. The empty level in the filled band, called a hole, has properties that are those of a free electron with a positive charge; the hole and the electron can be bound together, and will have a series of energy states similar to those of an electron and proton when bound together as a hydrogen atom. The combination of an electron and a hole which are usually bound in the configuration of lowest energy, is called an exciton, and the formation of an exciton will be revealed by appropriate absorption bands. Excited states have been observed in the alkali metal azides NaN_3, KN_3, RbN_3, and CsN_3; and at liquid helium temperatures, hydrogen exciton bands corresponding to quantum numbers, n, of 1, 2, and 3 have been resolved [3]. For example, with KN_3 these are at 1895 Å for $n=1$; 1545 Å for $n=2$; 1490 Å for $n=3$. Exciton bands have also been observed with thallous azide [4] and with silver azide [4, 5] (figures 1 and 7). Decomposition, by light of a wavelength corresponding to the exciton bands, involves, in the case of the alkali metal azides, the formation of F centres, that is, crystal defects in which an electron is trapped in a negative-ion vacancy, a point from

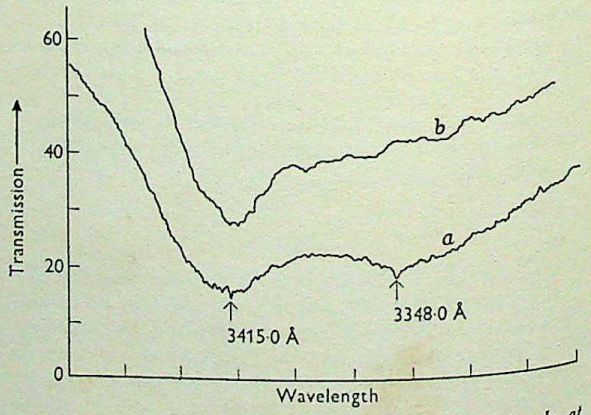

FIGURE 1 – *Microdensitometer trace of exciton bands at 3415 Å and 3348 Å in a thin evaporated film of thallous azide at liquid nitrogen temperature. Curve* (a) *for thin film* 0·14 μ *thick.*

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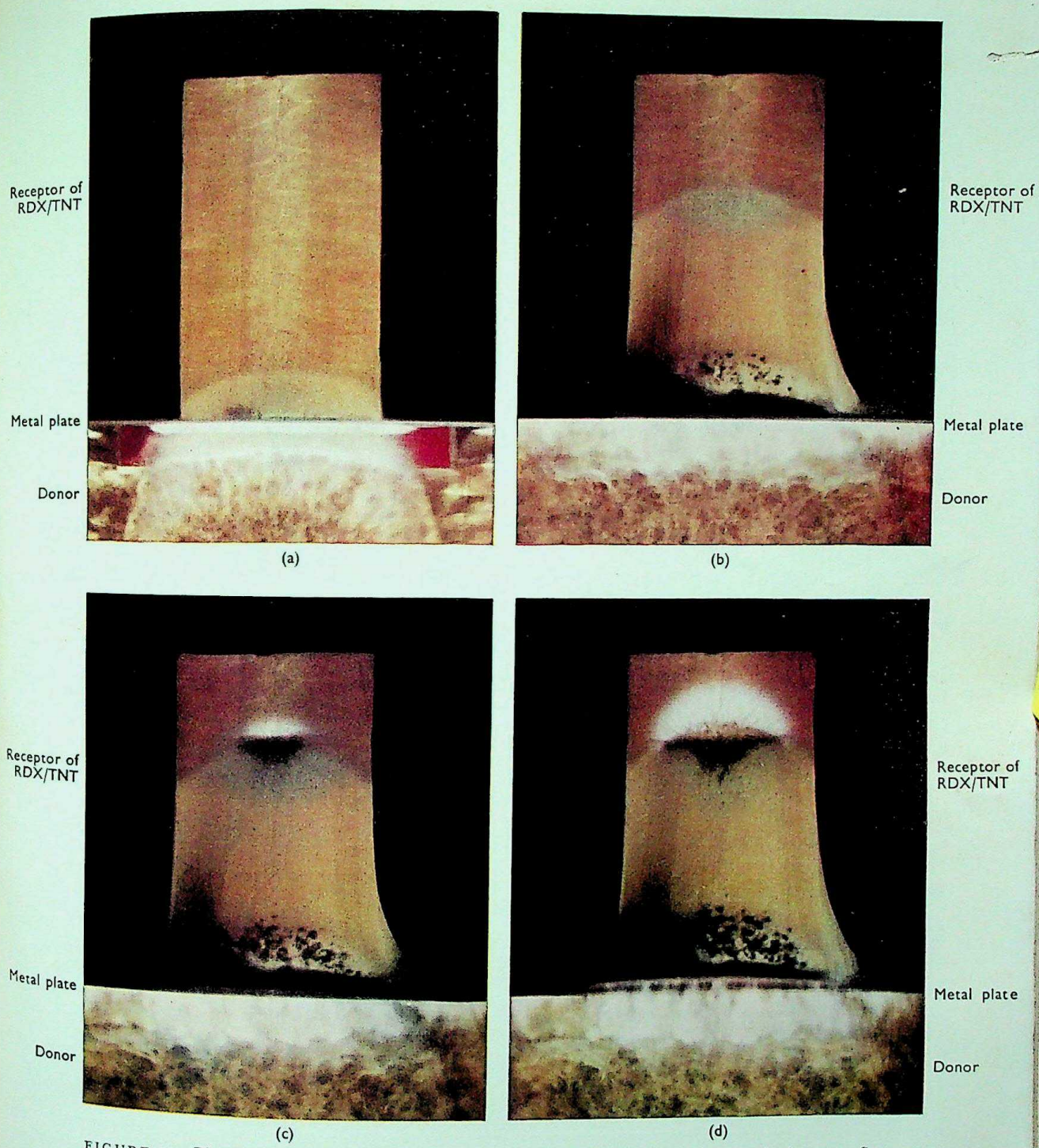

FIGURE 2 – *The detonation of a hemicylinder of* 60/40 *RDX/TNT by an intense shock wave. Sequence obtained on a camera with a framing rate of* 500 000 *per second, illuminated by an argon flash bomb (about* 40×10^6 *cp for* 100 μ*sec). The hemispherical receptor charge is* 6 *in. by* $3\frac{1}{2}$ *in., with the flat face towards the camera. The shock wave enters the receptor charge at the bottom of the picture. The position of the shock wave is clear in figures* 2(a) *and* (b). *Detonation commences, figure* 2(c), *well away from the surface where the shock wave originally entered.*

(Figures 2 and 6 supplied by Dr E. G. Whitbread and published with the permission of the Director, Ministry of Aviation, E.R.D.E., Waltham Abbey, Essex.)

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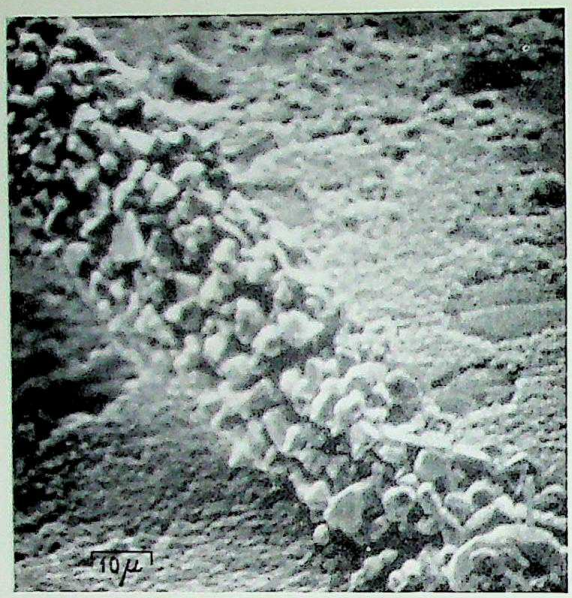


FIGURE 3 – *Needle of silver azide after heating, showing break-up into small blocks that then continue to decompose. Picture obtained using scanning electron-microscope.*

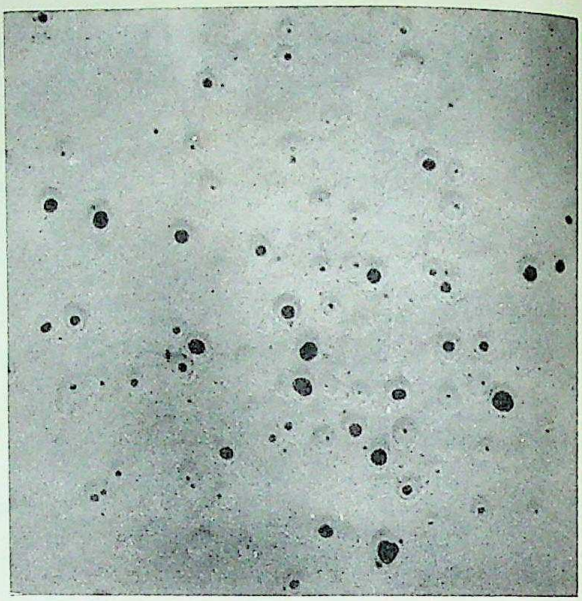
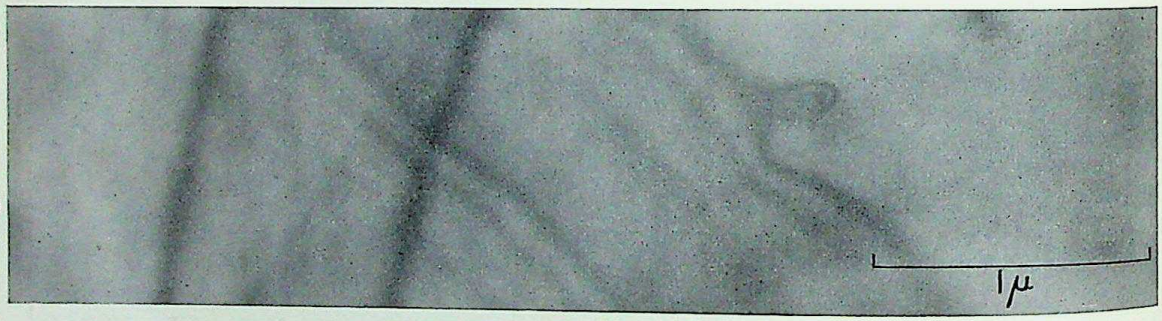
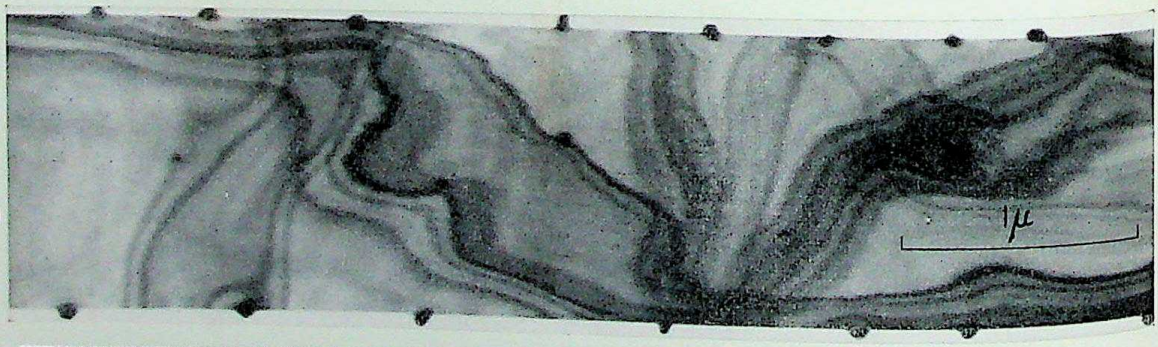


FIGURE 4 – *Electron micrographs showing the kind of silver nuclei formed on the surface of silver azide during successive early stages of thermal decomposition. Crystal heated for 7 minutes at 120° C.* (× 300 000)

(a)



(b)



(c)

FIGURE 5 – (a) *Transmission electron-micrograph of undecomposed silver cyanamide crystal, about 200 Å thick. The only visible details are the Bragg extinction contours arising from the slight elastic distortion of the lattice.* (b) *Crystal in early stages of thermal decomposition. A layer of the crystal extending from the surface to a depth of about 15 Å decomposes. The silver produced diffuses over the crystal surface and forms a few opaque specks at the edges.* (c) *Crystal irradiated with uranium fission fragments (neutron flux density of* $4{\cdot}3 \times 10^{15}$ *n vt). Each light area is a tunnel where material has been decomposed by a fragment. The mean track diameter is 120 Å. The dark areas are* [illegible]

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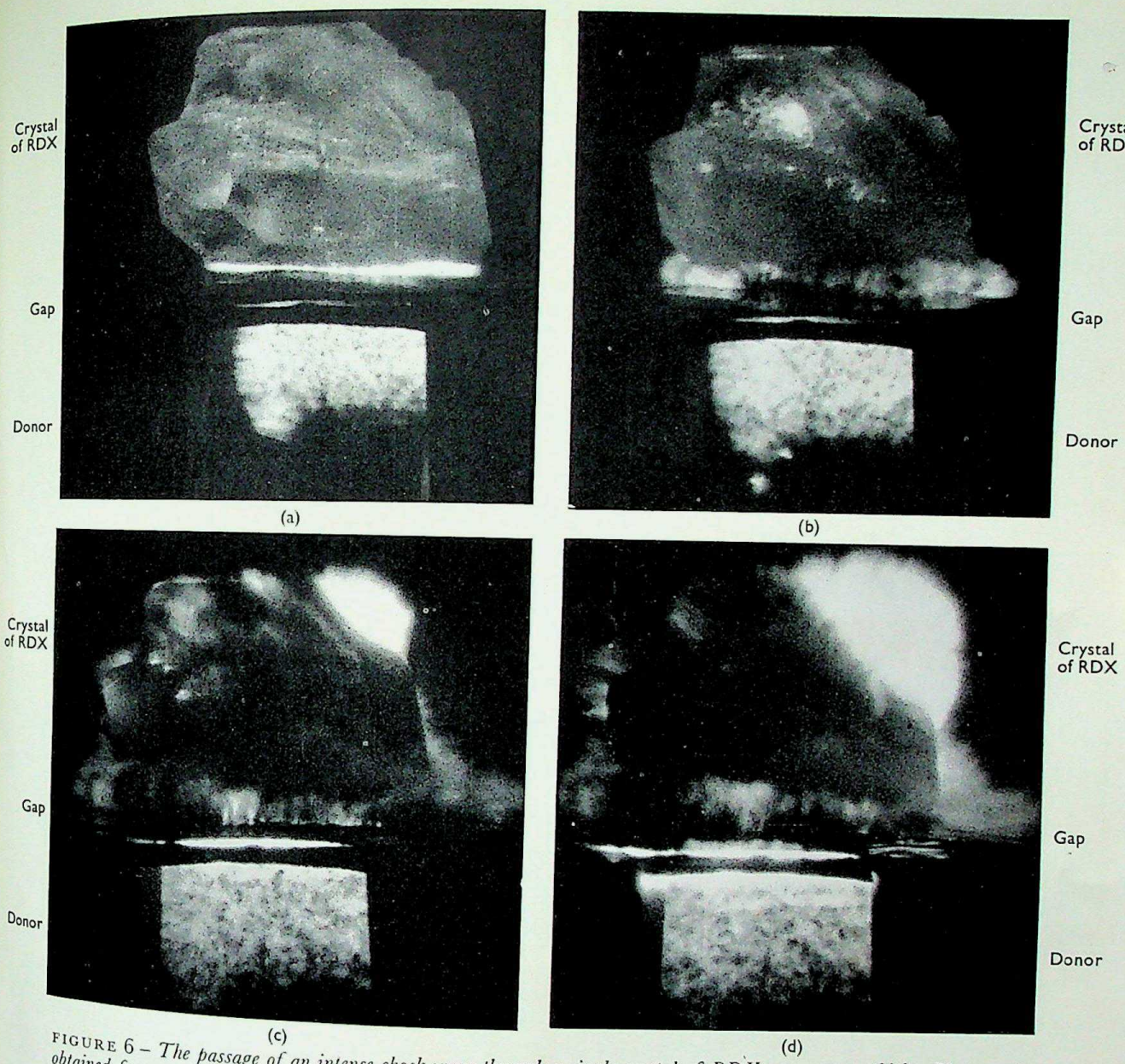

FIGURE 6 – *The passage of an intense shock wave through a single crystal of RDX, ca. 2·5 cm thick. Photographs obtained from a sequence using a high-speed framing camera operated at 1 200 000 frames per second. The crystal remains intact for over 5 μsec, then detonation starts, figure 6(c) at the far end of the crystal.*

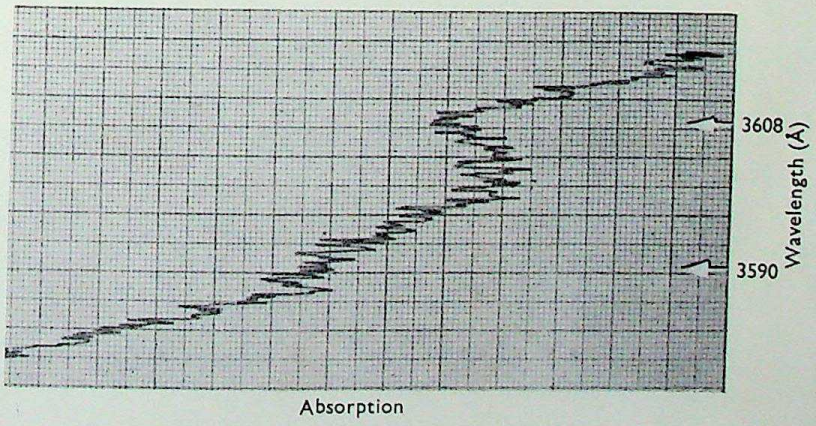

FIGURE 7 – *Microdensitometer trace of exciton bands at 3590 Å and 3608 Å in a thin crystal (6 μ thick) of silver azide at liquid nitrogen temperature.*

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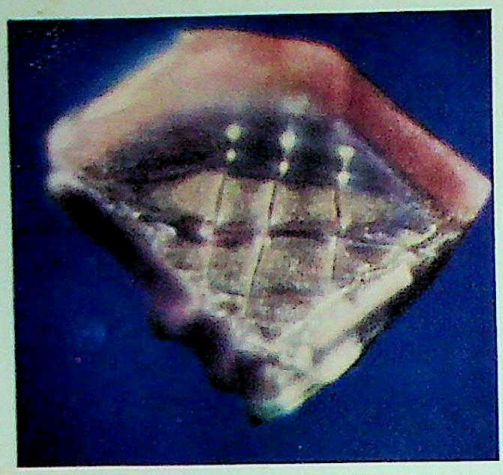


FIGURE 8 – *Crystal of cadmium azide after heating at 317° C, showing colouring of crystal and break-up along crystallographic planes.* (×600)

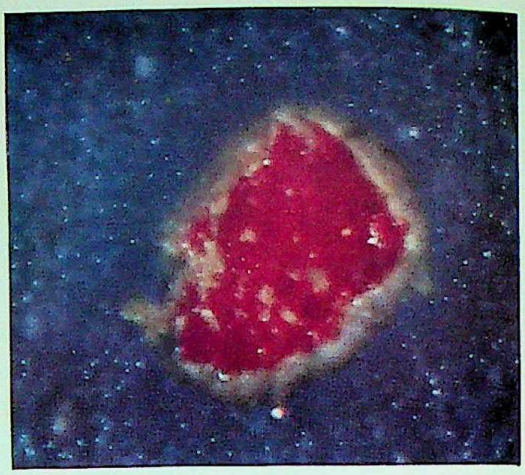
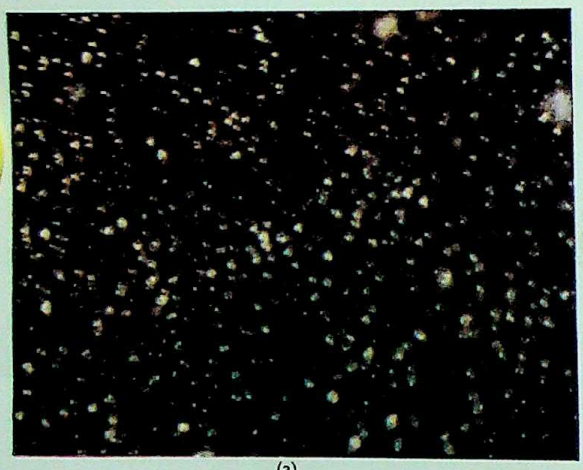
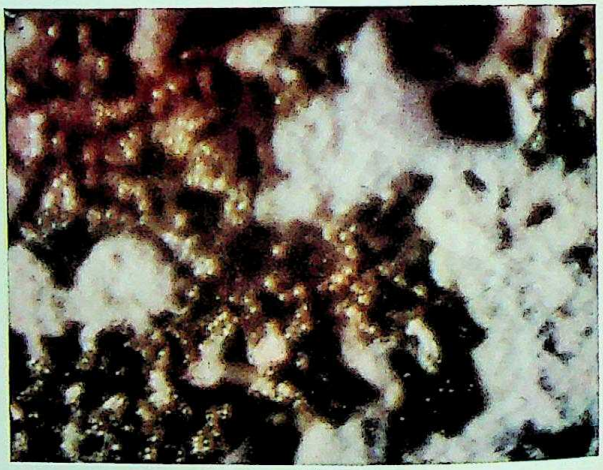


FIGURE 9 – *Surface of a pellet of silver azide containing a particle of mercuric iodide and flashed below the critical energy, showing smaller relative decomposition in the vicinity of the HgI_2 particle.* (×20)



(a)



(b)

FIGURE 10 – (a) *Surface of a pellet of silver azide flashed just below the critical energy for explosion, showing extensive decomposition over the whole surface* (×40). (b) *Surface of a pellet of silver azide containing a non-uniform distribution of gold particles flashed below the critical energy, showing the enhanced decomposition in the neighbourhood of the gold particles relative to the remainder of the surface* (×40).

which a negative ion is missing [6]. For the heavy-metal azides, such as thallous azide and silver azide, photochemical decomposition results from the thermal dissociation of optically formed excitons [2, 4, 5]:

$$N_3^- + h\nu = N_3^{-*}$$

Formation of an exciton.

$$N_3^{-*} \xrightarrow{kT} N_3 + e$$

Thermal dissociation of exciton and formation of positive hole and electron in the conduction band.

If light of wavelength corresponding to high absorption-coefficients ($> 10^5 cm^{-1}$) is used, there is the possibility of the azide ions dividing in this way:

$$N_3^- + h\nu = N_3 + e$$

In the case of azides and acetylides formed from the transition metals such as nickel and cobalt, the situation is a little more complicated. Here we have co-ordination complexes; ligand-field theory, that is, the study of the effect of the environment of anions on the energies of the *d*-orbitals of the cation, has proved valuable in discussing the optical and magnetic properties, and the stability, of these solids [7, 19].

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We have so far considered only the ionic azides from KN_3 to CuN_3. If the ionization potential of the metal is too high for the formation of an ionic bond, a covalent solid is formed. There is now direct bonding of the metal to the nitrogen atom of the azide group, and instead of the azide group being a linear symmetrical triatomic group, as in the azide ion, there are now differences in the N–N bond-lengths. Decomposition in the solid develops in a different way [4] and involves bond fission as the initial step:

$$M-N\text{——}N-N \longrightarrow M-N+N_2$$

Mercurous azide and the organic azides are solids of this type. The stability of these azides is very different from that of the 'ionic' azides. In general, the covalent solids are more stable to heat and light, and the order of stability is $KN_3 > TlN_3 > AgN_3 > CuN_3 < Hg_2(N_3)_2 <$ organic azides. Each azide must, however, be considered individually, and solid hydrazoic acid, for instance, is a covalent solid that is reported to be very unstable, although it does not seem to have been studied in great detail.

Further information about the slow decomposition of solids can be obtained by microscope studies using optical and electron microscopy (both transmission and scanning) and electron diffraction. It has been shown [8], for example, that crystals colour and break along crystallographic planes during decomposition (figure 8). In the case of silver azide, numerous small fragments about 1 μ across are formed (figure 3). In this way a large surface area is created during decomposition. This phenomenon is also important in the propagation of explosions and in low-velocity detonations. A detailed examination under the electron microscope has been made of the formation of metallic nuclei on single crystals of the azides [8]. In the case of silver azide, for example, the metal nuclei are formed at the surface of the crystal (figure 4). The evidence available at present shows that surface decomposition is more important than that occurring at defects within the crystal, although we do have evidence of internal decomposition causing cracking, so that fracture of the crystal may take place. J. W. Mitchell and his co-workers [9] have shown that, for a non-explosive crystal like silver bromide, the metal can be liberated at defects within. Even for an explosive solid such as silver cyanamide, which resembles silver azide, the silver is formed initially as a surface reaction. This silver is mobile and migrates to the edge of the crystal to form metallic nuclei [10] (figures 5(*a*), (*b*), (*c*)). It is clear that defects and dislocations will play some part in the decompositions. Dislocations in ionic crystals may be charged, and they may also be seats of reaction. The experiments made so far suggest, however, that with good crystals the surface reaction is of major importance in the early stages of the decomposition. Since the extent of decomposition prior to explosion is small, the surface reaction will also be important in the explosion process. Most of the work has been on the azides, but other compounds, such as the fulminates, are being studied in the same way. An interesting compound which has been studied in detail and which decomposes either by an electron-transfer mechanism or by a proton-transfer mechanism, depending on temperature, is ammonium perchlorate [18].

The work described in this section has been concerned mainly with the simpler azides. We would of course like to know more about the stability of such materials as lead azide, lead styphnate, and mercury fulminate. These solids are more complicated in both structure and optical properties. Nevertheless, it should be possible to study them in the same way as silver azide.

INITIATION OF EXPLOSION

Initiation by heat

One way to initiate an explosion is to use heat. Crystals can be placed in a furnace or heated in other ways, for example by infra-red radiation or by radio-frequency waves or microwaves. The three parameters that have to be specified for explosion are the thickness of the crystal, the temperature, and the time of application of the heat pulse. For a given thickness of crystal, the temperature required to produce an explosion increases as the duration of the heat pulse decreases. For example, with materials such as pentaerythritol tetranitrate (P.E.T.N.) the measured time to explosion is 10 milliseconds at 300° C and 50 microseconds at 550° C [11]. In this experiment, the solid is enclosed in a thin metallic needle which is heated by the rapid discharge of a capacitor charged to a very high voltage.

In order to produce an explosion, a 'hot spot' needs to be formed. For solids such as the azides and the organic nitrates, which decompose with the liberation of a good deal of heat, the radius of the hot spot must be greater than a certain critical value, which may be about 10^{-4} cm. The temperature should be in the region of 500° C and

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the lifetime about 10^{-4}–10^{-5} second. For a number of solids, the hot spot must be molten. Once such a hot spot is formed, the growth of reaction to an explosion of macroscopic size depends on thermal factors, such as the activation energies of decomposition, heat of decomposition, thermal conductivity, and specific heat. Heat is generated inside the hot spot at a faster rate than heat is lost by self-heating and by conduction. The theory both of the initiation and of the growth processes for thermal explosion is fairly well understood. The general equation for heat balance that determines the growth of the hot spot is

$$\underset{\text{Self-heating}}{\rho C_v \frac{\partial T}{\partial t}} = \underset{\text{Heat gained by conduction}}{K\nabla^2 T} + \underset{\text{Heat generated by chemical reaction}}{\rho Q A e^{-E/RT}}$$

This corresponds to a hot spot of radius a and temperature T, where the density of material is ρ, thermal conductivity K, specific heat C_v, and the bulk temperature T_0. The heat of decomposition is Q calories per gram, r is the radial distance from the hot spot, E the activation energy, and A a constant. The boundary conditions are $T = T_0$ for $r > a$, and $T = T_1$ for $r < a$, when $t = 0$. The exact solution of the above equation is difficult to obtain. Various approximations have been made, and numerical integration methods have also been used. These have given values for critical hot-spot sizes as a function of temperature, and have also given a relation between induction periods for explosion and temperature. One of the useful relations obtained by Frank-Kamenetsky gives values for the critical radius r of material required for explosion in terms of the heat of reaction and activation energy. For a spherical system:

$$\delta = 3{\cdot}32 = \frac{Q}{K}\frac{E}{RT_0^2} r^2 A \exp(-E/RT_0)$$

T_0 is the exterior surface temperature and T_1 the temperature in the interior of the sphere. When δ exceeds 3·32, self-heating leading to explosion will occur. The temperature rise inside the hot spot necessary for explosion is given approximately by

$$T_1 - T_0 = \Delta T \simeq RT_0^2/E$$

A simple calculation shows that the temperature rise ΔT necessary for explosion to occur is in fact very small—it may be as low as 10° C.

Initiation by shock

Experiments suggest that the initiation of explosion by shock is usually thermal in origin [1]. Explosions may be initiated by impact, friction, shock tubes, ultrasonic vibrations, or flying particles that can be as small as a micron in radius and have velocities higher than 1 km per sec. Shock waves have also been used. These range from gentle shocks to intense shock waves, where the peak pressure can be in the region of 100 kbar (100 000 atmospheres).

In the case of the initiation of explosion by gentle impact, it seems that the mechanical energy of the impact must first of all be degraded into heat in a localized region of the solid or liquid; explosion then grows from this hot spot by a normal thermal process. If tiny gas bubbles or voids are trapped inside the liquid or solid, then, during impact, the gas is heated by adiabatic compression. If a small bubble is present in nitroglycerine for example, explosion may be initiated by the gentle impact of a few hundred grams dropping a centimetre or so. If bubbles are absent, the necessary impact energy will be two orders of magnitude higher.

For an ideal gas, the relation for the temperature rise is given by

$$T_2 = T_1(P_2/P_1)^{(\gamma-1)/\gamma}$$

where T_1, T_2 are the initial and final temperatures in the gas bubble, P_2/P_1 the compression ratio, and γ the ratio of the specific heats. Temperatures of about 500–1000° C are generated for times of about 10^{-5}–10^{-4} seconds. The compression ratio is then about 20 : 1. The quantity of heat localized in the gas bubble during impact is small, and for nitroglycerine it can be about 10^{-8} cal and still produce an explosion. These values actually correspond to upper limits, as the compression is not truly adiabatic, and vapour and fine particles of material must also be present inside the gas bubble. If grit is present, frictional heating can occur at localized points; the maximum temperature reached by the grit particle is limited by its melting point. Provided that this melting point is higher than the explosion temperature corresponding to the time of application of the heat pulse (about 10^{-4} sec), an explosion will develop from this hot spot. These temperatures are in the region of 500° C. Experiment shows that the property of the grit most important in determining the incidence of explosion is the melting point, although the size, shape, hardness, and plastic properties must all be considered. For example, if a striker is used with a sharp point, plastic flow and heating can occur at the tip and the temperature rise may be sufficient to initiate

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an explosion. For liquids, in the absence of air pockets or grit particles, explosions are obtained only at high energies of impact, and here viscous heating of the liquid as it flows between the colliding surfaces can be sufficient to cause an explosion.

The results obtained using other methods of shock initiation can also be interpreted by a thermal mechanism. For example, when ultrasonic vibrations are used, we get cavitation and the collapse of bubbles, giving rise to thermal effects; here again other factors, such as electrical effects and free-radical formation, must be considered.

It must be admitted, however, that the detailed mechanism of the shock initiation of explosion still needs some clarification. Some Russian investigators have proposed that the latent heat of fusion is an important factor in initiation of explosion by impact; the importance of visco-plastic flow has also been emphasized. Other workers consider that true 'tribochemical' reactions, that is, chemical reactions in which physical shock as such plays a part, are important in shock initiation. The great difficulty is in the design of suitable experiments to distinguish between the mechanisms that have been suggested. The thermal mechanism, however, does seem to cover most of the phenomena that have been observed during initiation by gentle shock.

A considerable number of papers have been published [11, 12] on the effect of intense shock waves on liquids and solids. The results are beginning to fall into a pattern. It seems that if we have a homogeneous system—for example a liquid without gas bubbles or a solid without defects, imperfections, or voids—such a system is very insensitive to shocks of very high intensity. Thus shock waves with a velocity of 3000–3500 m per sec and shock pressures of about 40 kbar may not detonate bubble-free nitroglycerine [11]. For nitromethane, pressures of about 85 kbar are insufficient to initiate detonation in a homogeneous system. Similarly, single crystals of RDX (cyclotrimethylene trinitramine) do not detonate during the passage of a shock wave; detonation starts from the end of the crystal remote from the source of the shock [13]. Figure 6 shows a crystal of RDX subjected to a shock wave. It preserves its form for 5 μsec; after this a detonation, probably due to a dust explosion, starts at the end of the crystal. The shock wave itself is sufficient to raise the temperature by only a few hundred degrees, and this is below the thermal ignition temperature. If the shock wave is very intense (about 100 kbar), we may calculate that the temperature rise may be higher than 400° C, and a thermal explosion will result, leading to detonation.

With a non-homogeneous liquid or solid the situation is different. If we compress powder to a pellet in which voids are present, or if we have solid foreign particles in the pellet, explosion and detonation can occur with moderately intense shock waves. Figure 2 shows how detonation in a solid pellet begins at a region well away from the surface where the shock wave originally entered. The shock wave starts a reaction in the solid that is sufficiently fast to accelerate the shock wave. This builds up into a detonation after a time interval that depends on the surface area of the explosive material. Again, a thermal mechanism can be proposed for the initiation of explosion and detonation. Adiabatic compression of gas pockets (the value of γ here probably corresponds to that for monatomic gases), and reflection of shock waves from inhomogeneities in the liquid or solid, which can result in the intersection and reinforcement of the shock waves, can cause a high local temperature; these are possible sources of initiation. In other words, for a non-homogeneous solid there is a distortion of the plane shock wave leading to localized regions of very high pressure and therefore very high temperature. As the reaction rate increases exponentially with temperature, the effect on the overall decomposition rate is higher than for the undisturbed wave.

One idea which has been put forward many times is the possibility of the direct rupture of bonds by the shock wave. A shock wave moving with a velocity of 10 000 m/sec takes 10^{-14} sec to traverse 1 Å, and this is of the same order as the vibration periods for unexcited molecules. Longitudinal rupture of the bonds is therefore a possibility. Direct rupture of bonds under shear has also been postulated. This suggestion must be kept in mind, but there is at present no real evidence for this mechanism in 'normal' liquids and solids, where the molecules are fairly small.

Initiation by light

There has been a considerable amount of study of the initiation of explosion by light, following the pioneer work of J. Eggert and his colleagues [14]. Light sources are now available that can give out high intensities in short times, up to 10 000 joules in 10^{-3} sec. The simplest technique is to discharge a bank of condensers between metal electrodes in argon, using suitable optical filters

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to select the right wavelength region and to eliminate infra-red radiation. The light is allowed to fall on the solid to be studied, and this is absorbed in a very thin layer near the surface if the wavelength approaches the optical absorption edge. The critical amount of light-energy required for explosion can be determined within fairly close limits [1, 14, 15]. Crystals are not damaged during this process, and for this reason the method is not subject to the uncertainties inherent in shock initiation of explosion. Some typical relative results obtained with the azides are given in the table.

These results show that our general conclusions about the stability of solids apply to explosion initiated by light [2, 15]. The order of stability to light is thus $KN_3 > TlN_3 > AgN_3 > CuN_3 < Hg_2(N_3)_2$. The initial step is a direct photochemical decomposition, and this is followed by a growth to explosion that depends on thermal factors. The critical conditions that determine explosion can be obtained approximately by solving the simplified equation for heat flow:

$$C\rho \frac{\partial T}{\partial t} = K \frac{\partial^2 T}{\partial z^2} + \rho Q A e^{-E/RT} + \alpha E_0 e^{-\alpha z}$$

Here α is the absorption coefficient, z the coordinate giving the thickness of the crystal, and E_0 the incident light flux per second [16]. Eggert [14] considers only the possibility of thermal explosions in initiation by light.

The ionic azides can also be sensitized or desensitized by the introduction of foreign anions [15, 20], and by solid impurities of the right kind. Metal particles such as gold or silver sensitize the azides [2] (figure 10), while electron donors such as HgI_2 desensitize them [15] (figure 9). These results can be interpreted in terms of the electronic processes that occur in the solid during decomposition. For example, it is postulated that metal particles adsorb positive ions and form electron traps, and thus enhance the decomposition; with HgI_2 the reverse is true, and there is an increase in electron density.

Covalent organic azides and nitrates, on the other hand, are not exploded by an intense light flash when in the solid state, even at temperatures close to the melting point. Molten materials such as P.E.T.N. can, however, explode at reasonable light-energies. With these materials, it is necessary to consider the mobility and the reactivity of the free radicals which are formed as intermediates in the reaction.

Initiation by fission fragments and ionizing radiations

The inorganic azides decompose and explode more easily as the wavelength of irradiating light decreases. However, no explosions have been observed with very short wavelength light or with soft X-rays and γ-rays. Defects, interstitials, and colour centres may be produced, as well as decomposition, but these do not lead to explosion. This is undoubtedly because the penetration distance into the crystal is larger than for long-wavelength light, and the intensities available are not high enough to cause sufficient reaction in a

Ignition behaviour of some inorganic azides

Compound	Melting point (°C)	Heat of decomposition to metal (kcal/mole)	Thermal ignition temperature (°C)	Optical ignition energy (joule)	Detonation velocity (m/sec) in thin film ca. 0·1 mm thick
KN_3	343	0	—	—	—
TlN_3 ..	334	56	475	92	1480
AgN_3 ..	250	74	475	39	2170
CuN_3 ..	205	67	210	12	2720
$Hg_2(N_3)_2$ ..	210	141	400	164	2350
$Hg(N_3)_2$ ..	190	—	460	90	ca. 3000
$Pb(N_3)_2$ ..	—	115	370	29	2300

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short time. Even large high-energy nuclear particles such as α-particles and fission fragments do not bring about the explosion of the azides under the conditions of our experiments. In the case of fission fragments of known energy, it is possible to estimate the extent of decomposition (radius and length) as the fission fragment ploughs through the crystal. A typical track in silver cyanamide [10], for example, is shown in figure 5. Silver cyanamide is a compound that resembles silver azide. The diameter of the hot spot or hot cylinder in these experiments is about 120 Å. This is below the critical size necessary for explosion, and again the results agree with values obtained by other means. The calculations suggest that for a high concentration of fission fragments entering a crystal, an explosion might occur if a few tracks intersected within a very short time. Up to the present, the only material that explodes during irradiation by α-particles and fission fragments is nitrogen iodide [8]. This, however, is essentially a surface reaction in which the ammonia, which is normally linked to the NI_3, is removed, leaving behind the unstable NI_3, which decomposes spontaneously. A considerable amount of work remains to be done on radiation effects in explosive solids, investigating, for example, the effects of tracks that intersect.

GROWTH OF EXPLOSION TO DETONATION

Once an explosion is initiated in a single crystal or in a thin film, about 0·1 mm thick, of an explosive material, this explosion can grow and develop into a low-velocity detonation; low-velocity detonations may be initiated by heat, light, or shock waves. In order to follow the transition from explosion to detonation, and for the measurement of low-velocity detonation, a variety of devices may be used. These include drum cameras, framing cameras, image-convertor tubes, and fast-recording spiral-base oscilloscopes. With this apparatus, detonation velocities as a function of film thickness and intensity of initiation may be measured. Detonation velocities in the region 1000–3000 m per sec have been recorded. It is true to say that there is at present no satisfactory theory to account for the phenomenon of low-velocity detonation. It is necessary to explain, for example, why the low-velocity detonation is stable, and why a considerable amount of material is left unconsumed after the detonation wave has passed. Low-velocity detonations in the region of 1000–2000 m per sec are stable, and propagate over considerable lengths. It is possible to build up from a low-velocity detonation to a high-velocity detonation, but the reverse process will not normally occur. Instead, the detonation dies out if the high-velocity detonation degenerates. The shock parameters (pressure, temperature, and zone length) in low-velocity detonation are known fairly accurately, and simple calculation shows that, for a homogeneous solid, the temperature rise due to compression is negligible. It is, therefore, necessary to look for another process that is able to support the reactions necessary for low-velocity detonation. It appears that for propagation of low-velocity detonation it is necessary to have defects of one kind or another. These can be small gas pockets. Factors that could aid the propagation of the low-velocity detonation are compression of gas pockets ahead of the reaction zone, break-up of the solid to expose large surface areas, reflection and intersection of shock waves, and frictional heating of rubbing particles.

Calculations based on the hydrodynamic theory of detonation have been made by R. Schall [17]. It is assumed in his calculation that incomplete decomposition of the solid occurs. This leads to the result that it should be possible to produce stable low-velocity detonations over a fairly wide range of velocities. Refinements of this theory include decomposition in steps [16], and this gives results that are more reasonable than those of the basic theory. The hydrodynamic theory, of course, tells us nothing about the processes that occur during detonation.

In the case of azides, a number of points need clarification. Firstly, the values obtained for detonation velocities of a number of azides spread as thin films are not very different (see table). That is to say, the magnitude of the low-velocity detonation is not very structure-sensitive. Secondly, detonation has been observed in very thin needles (less than 0·1 mm diameter) of, for example, silver azide, where the diameter is about the same as or smaller than the thickness of the reaction zone. As the thickness is reduced still further, the velocity decreases until finally a burning region is obtained. This raises the question as to whether burning and detonation are similar processes. Recently, Boddington in this laboratory has made a study with a microsecond framing camera of the shock initiation of explosive single crystals as needles and as films about 0·1 mm thick. The low-velocity explosion regime is in fact a very rapid deflagration for the primary explosives. The deflagration front consumes all the explosive and is preceded by a weak inert shock

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wave of greater velocity. The homogeneity of a crystal with a perfect habit makes it extremely insensitive towards shocks. When there are imperfections of habit present with dimensions as small as 10 μ, then these can act as sites of high shock-sensitivity.

All the experiments considered above involve the use of thin films that are only moderately confined. If, however, the thin film of explosive is heavily confined, for example in lead tubes, it is possible to propagate high-velocity detonations over very large distances.

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Parthenogenesis in the Phasmidae

J. BERGERARD

Some species of Phasmids are insects that can reproduce either sexually or parthenogenetically, in which case only females are produced. This article discusses the mechanisms by which this parthenogenesis is established, and it also describes experiments by which the sex of the female insects can be altered, giving in some cases insects of intermediate sex.

Phasmids—stick insects—belong to the same family as grasshoppers and locusts. They have elongated twig-shaped bodies that frequently enable them to pass unnoticed in foliage. In addition, the Phasmids change colour in response to their surroundings. Such changes are extremely rare among insects; in this case hormones produce a migration of the pigments in the epidermal cells [8].

Although in Australia, for example, Phasmids are so numerous as to menace the eucalyptus tree, there are only a few species in temperate regions, and they contain relatively few insects. This, and their unusual behaviour, led to something of a scientific craze about the beginning of the century, when many laboratories began to breed them from seed-like eggs brought or sent back by missions, especially from South-east Asia. It was in this way that *Carausius morosus*, which originated from India, became a classical laboratory animal.

The insects are easily bred. They are so easily reared, indeed, that many children rear them as they do silkworms. The only disadvantage is that they mature slowly, most of the species taking a year to complete one reproductive cycle. Many feed on the leaves of trees or bushes without showing any particular discrimination. *Carausius morosus*, for example, can be reared on ivy leaves just as successfully as on blackberry bushes. Certain species, however, appear to be more demanding. The giant of the group, *Eurycnema herculeana*, an adult female of which measures about 25 cm long, has so far been reared completely in our laboratory only on oak leaves. This necessitates adapting the breeding of the insect to the annual growth cycle of the tree and prevents the animal, despite its great size, from becoming wholly satisfactory for physiological research. Laboratory breeding has quickly revealed many rather odd characteristics of the reproduction of these insects compared with that of other animals.

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PARTHENOGENESIS

The various Phasmid species can be divided into two categories, one containing males and females that reproduce in the normal bisexual manner, the other containing only females that multiply by means of unlimited parthenogenesis. *Carausius morosus* is an example of this latter group. Thus far there is nothing very original, as many other animal groups show the same pattern. However, it has been found [5] that the females of bisexual Phasmid species can, when separated at an early age from males, lay eggs that will develop in the same way as those of parthenogenetic species. In the extreme case, females of certain species isolated in this way produce eggs that develop just as if they had been fertilized by a male. This happens, for example, with *Clitumnus extradentatus*, which originates from the Annam coast and has been bred in our laboratory for fifteen years or so (figure 1). Here, parthenogenesis is an alternative to sexual reproduction—it is facultative. The alternative, accidental parthenogenesis, is known in a number of other groups. In species showing facultative parthenogenesis the sole apparent function of the male is the propagation of other males and hence, probably to ensure that a number of insects remain heterozygous; the females alone can produce a fairly numerous progeny capable of indefinite perpetuation but composed solely of females.

In order to discuss instances of parthenogenesis, it is necessary to understand the cytological mechanism. In normal reproduction, the fertilized egg contains $2n$ chromosomes in its nucleus, n coming from the nucleus of the ovum and n from the fertilizing spermatozoon. Ova and spermatozoa originate from cells with $2n$ chromosomes,

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diploid cells, which in the course of two special divisions (maturation divisions) give four haploid cells, with *n* chromosomes. In most of the known instances of regular parthenogenesis the two maturation division are modified (most frequently being replaced by only one), producing a 2*n* ovum capable of developing without the aid of a spermatozoon. Parthenogenesis is then established in the species. The detailed mechanism will be discussed later.

Facultative parthenogenesis can cause unusual variations in geographical distribution. Over one part of its distribution area a species may be bisexual; over another, parthenogenetic. This situation may arise from 'accidental' causes, as seems to be the case with *Sipyloides sipylus*. In its original habitat (Malay archipelago, Sunda Isles) the species is bisexual; in Madagascar, where its introduction appears to be recent, only females occur. Elsewhere climatic factors can no doubt play a part. The species *Bacillus rossii* contains practically only females in the south of France, but both males and females in north Africa. In this instance we know that only one species is involved because the chromosome make-up is similar, and because females from the south of France have been successfully mated with males from north Africa. The resulting progeny of roughly equal numbers of males and females shows the effectiveness of the fertilization.

The problems are not always so simple, however, and where distribution areas overlap, one frequently finds two species, one bisexual, the other parthenogenetic, that are very close to each other morphologically. One example has been particularly well studied; in this, the species are *Leptynia hispanica* (Spain, Portugal, and the French Mediterranean coasts) and *Leptynia attenuata* (Spain, Portugal) [6]. The first multiplies only by parthenogenesis and is triploid; the second is bisexual and diploid, and reproduces sexually. This is a case of geographical parthenogenesis, therefore, with polyploidy of the parthenogenetic species.

The Phasmidae is obviously a particularly interesting group in which to study parthenogenesis, as it offers cases forming a more or less continuous bridge between the different types of parthenogenesis that are encountered only in a much more fragmentary manner in other groups of animals. In particular, it contains all the intermediaries between accidental parthenogenesis, where virgin eggs give a very small percentage of normal larval eclosions, and truly facultative parthenogenesis, where eggs have a percentage eclosion of 80–100. The different instances enable one to follow a probable line of evolution in the establishment of the geographical parthenogenetic breeds.

A large number of species originating from India [5] display an accidental parthenogenesis similar to that reported [3] for many Acridiidae species. In these, the unfertilized eggs show initial haploid development; this is followed by more or less complete diploidization, which results mainly in embryonic malformations, although a few per cent of eggs hatch. This type of accidental parthenogenesis does not establish itself in nature.

In other species, for example *Menexenus semiarmatus* [5] (44 per cent eclosions) or *Clitumnus extradentatus* [1] (60 per cent eclosions), virgin eggs develop more regularly. In the latter case it is possible to breed stocks showing truly facultative parthenogenesis in which almost all the virgin eggs hatch. With an increase in the percentage of virgin eggs that develop normally, the diploidization of the segmentation nuclei becomes more regular. A species such as this is potentially able to form parthenogenetic breeds.

Such breeds may be the result of fortuitous isolation of the females, as would be the case with *Sipyloides sipylus* in Madagascar; they may also result from the influence of climatic factors. In the Phasmids the male is much more delicate than the female. Many authors have recorded very abnormal proportions of the sexes, and it has been proved that in stocks of *Clitumnus extradentatus* the proportion of males, 50 per cent at eclosion, falls to about 25–30 per cent in populations of old larvae or young adults.

An example of a parthenogenetic breed formed in this way is *Bacillus rossii*. The males have disappeared in the northern part of its distribution area, which suggests that the reason is the change of climate. However, it is still possible to fertilize these normally parthenogenetic females with males taken from more southerly populations. This, therefore, is the least advanced case of geographic parthenogenesis imaginable.

There are other groups of animals that show a more extreme form. These contain two breeds, one generally occurring nearer the equator and containing both males and females, the other parthenogenetic. The parthenogenetic females are polyploid and cannot be fertilized. Examples are the woodlouse *Trichoniscus elizabethae* studied by Vandel, the Psychid butterfly *Solenobia triquetrella* described by Seiler, and the many weevil species studied by Suomalainen. These breeds do not show any intermediate forms that can fertilize

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one another, so it is difficult to know how they have become established.

Certain Phasmid species have reached this stage of evolution. We have already referred to the case of the two similar species *Leptynia attenuata*—a bisexual diploid, and *L. hispanica*, parthenogenetic and triploid. No study has been made unfortunately of the regulation of the number of chromosomes in this second species; it is probably based on anomalies in the maturation divisions. It is difficult to imagine a normal chromatic reduction in a triploid organism.

This example more or less marks the end of the evolution of parthenogenesis in the Phasmidae, the actual end being the disappearance of the bisexual breed. It is difficult to allot an exact position, in terms of the evolution of parthenogenesis, to the species, *Carausius morosus*, exhibiting constant parthenogenesis. Species of the same genus are known with 42 chromosomes [5]. *Carausius morosus* has 64 chromosomes [3] and could pass for a triploid; but the artificially produced males that will be discussed later have the same number of chromosomes as the females and pair off these chromosomes as 32 bivalents at meiosis. The Phasmids appear to constitute the easiest group in the animal kingdom in which to follow the evolution of parthenogenesis because of the multiplicity and diversity of cases they present.

CYTOLOGICAL MECHANISM OF PARTHENOGENESIS IN THE PHASMIDAE

There are a number of problems concerned with this mechanism. In facultatively parthenogenetic species, for example, it must be fairly close to the normal mechanism if the egg is to retain the possibility of natural fertilization, and at the same time fairly regular if viable larvae are to be the outcome in most cases. I have been studying the cytological mechanism in the species *Clitumnus extradentatus* (figure 1), which we have been breeding in our laboratory for fifteen years [1]. In this species all the virgin females lay a large number of eggs, of which about 60 per cent hatch. Observation of virgin females isolated individually, and of their progeny, has shown that one can isolate lines with a very high percentage, up to 95 per cent, of parthenogenetic eclosions; other lines give much smaller and more variable percentages. The ability to reproduce parthenogenetically persists in the different lines for more than ten successive generations in each of the lines studied. The embryonic development of the virgin eggs in lines with a high percentage of parthenogenetic eclosion will be seen to be extremely slow to start with. The total duration, 57 days, is much longer than the 45 days needed by eggs fertilized normally. The part of the development phase that is slowed down in this way may cover the development of the embryo to a more or less advanced stage. This stage differs in different stocks. It is capable, however, of extending to the stage of a metameric germ-band in which the nervous system is already fully developed.

These slowly developing embryos consist at first solely of haploid cells (n chromosomes); then of a mixture of haploid and diploid cells ($2n$ chromosomes); and finally, when their rate of development overtakes that of embryos resulting from fertilized eggs, solely of diploid cells (figure 5, right). The essential mechanism of this chromosome regulation is revealed by the existence, in addition to normal haploid or diploid mitoses, of numerous blocked haploid mitoses similar to those blocked by, for example, colchicine. There are no degenerating pycnotic cells during the evolution of the embryo from the haploid to the diploid state. This shows that after a relatively long period of blockage, one of these blocked metaphase cells enters upon sets of interphase; the two chromosomes that have been kept together by their centromeres now find themselves grouped inside a single nucleus. Subsequently, such a cell multiplies in diploid fashion.

These facts explain the main characteristics of this facultative parthenogenesis. Firstly, the ovum continues to be fertilizable because it is haploid. Secondly, all the progeny are female because in this species, as in most of the Phasmidae, the chromosome mechanism for determining the sex is of the type XO in the male and XX in the female. All the ova therefore contain one X-chromosome and the spermatozoa either one X-chromosome or no heterochromosomes. On fertilization 50 per cent of the eggs will be XX and give females, 50 per cent XO and give males. The non-fertilized ovule is haploid with one X-chromosome and when the chromosomes double, the cells become diploid with 2X; the resulting embryo will therefore be female.

It should be noted that this method of chromosome regulation is not at all common in the animal kingdom among species in which parthenogenesis is well established. The only well-known case is that of the Psychid butterfly *Solenobia triquetrella* studied by Seiler [13, 14], and even in this case the doubling of the chromosomes is achieved rather by nuclear fusions taking place at the very

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beginning of the segmentation of the egg. On the other hand, an identical mechanism is at work in the accidental parthenogenesis of Orthopterae, in which, however, diploidization is only partial [3, 10].

This mechanism is possibly a stage in the evolution of the classical forms of parthenogenesis in which the two maturation divisions are modified. In addition, it reveals a possible link between the establishment of parthenogenesis and the realization of polyploid breeds. The doubling of the number of chromosomes in the haploid cells manifests a fundamental tendency towards polyploidization, which, acting on eggs already diploid, can give rise to individuals with $4n$ chromosomes. Triploid specimens could result from $2n$ ovules fertilized by an n spermatozoon, as in *Solenobia*. The establishment of polyploid breeds of course still depends on natural selection being in their favour.

SEX DIFFERENTIATION

The strange facts of parthenogenesis in the Phasmidae that have just been described are not the only elements of interest in the sexual biology of these animals. Most of the bisexual species exhibit very considerable sexual dimorphism (figure 1), and it has long been known that male or highly intersexed individuals sometimes appear accidentally in breeding stocks of constantly parthenogenetic species. These 'males' are generally very rare: for example one out of 1000–10 000 *Carausius morosus* bred in our laboratory.

Two different interpretations have been suggested: gynandromorphism and intersexuality [4, 9, 12, 15]. I have been able to show that sex differentiation in *Carausius morosus* is in fact dependent on external temperature. Eggs incubated during the first third of the embryo's life at 30°C (controls incubated at 23°C) give as many as 100 per cent males. The percentage is not comparable with the number of accidental males appearing spontaneously in breeding stocks [2].

The sexual dimorphism of the Phasmidae shows itself in the secondary sexual characteristics, in internal and external primary characteristics, and in the characteristics of the gonads. The most obvious secondary sexual characteristics are that the male is much smaller; his antennae are relatively much longer; his cuticle is smooth and waxy instead of finely granulated as in the female; and he has a red spot on the ventral surface of the thorax that is lacking in the female. The external primary characteristics are the external genital pieces. In the female the four valves of the ovipositor are covered by the subgenital plate. These valves derive from the eighth and ninth abdominal segments, and the sub-genital plate represents the sternite of the eighth segment. In the male the copulatory pieces, which are relatively simple, derive from an invagination situated between the ninth and tenth abdominal segments. They are covered over with a sub-genital plate representing the sternite of the ninth segment. The internal primary characteristics relate to the genital ducts. In the female the oviducts end originally (in the young larvae) on the ventral surface of the seventh and eighth abdominal segments. Later they are prolonged by a common oviduct forming a copulative and a laying chamber. Two accessory glands open into the copulative sac (figure 6a). In the male the spermiducts lead into a bundle of accessory glands that eventually open between the ninth and tenth abdominal segments: this pattern is shown throughout the whole larval and adult stage. The gonads in the male are two tubular testicles following directly from the spermiducts and extending from the third to the sixth abdominal ring (figure 6b). The gonad in the female is formed by ovarioles attached separately on the prolongation towards the anterior end of each of the oviducts; each ovary comprises twenty or so ovarioles.

The period of sensitivity to the higher temperature for the differentiation of these characteristics covers the first 30 days of the embryo's life; the total development period at 23°C is about 80 days. At the end of 30 days the embryo is already well formed and is starting dorsal closure in its posterior region. Shorter treatments may be given, the developing eggs being taken to 30°C for a week or fortnight within this period of sensitivity. In this way a whole range of intersexed specimens can be obtained, masculinity increasing with length of treatment (figures 3, 4).

It is strange that, as masculinity increases, the secondary sexual characteristics are the first to be affected, although these become differentiated very late in the life of the larva. The changes then affects the external genital pieces and the genital ducts. It is only animals that are very masculine in external appearance that possess either ovotestes or real testicles.

The easiest characteristics to interpret are the external genital pieces and the genital ducts in particular. Indeed, as has already been mentioned, the outlines of these organs are different in the two sexes, and intersexed specimens generally

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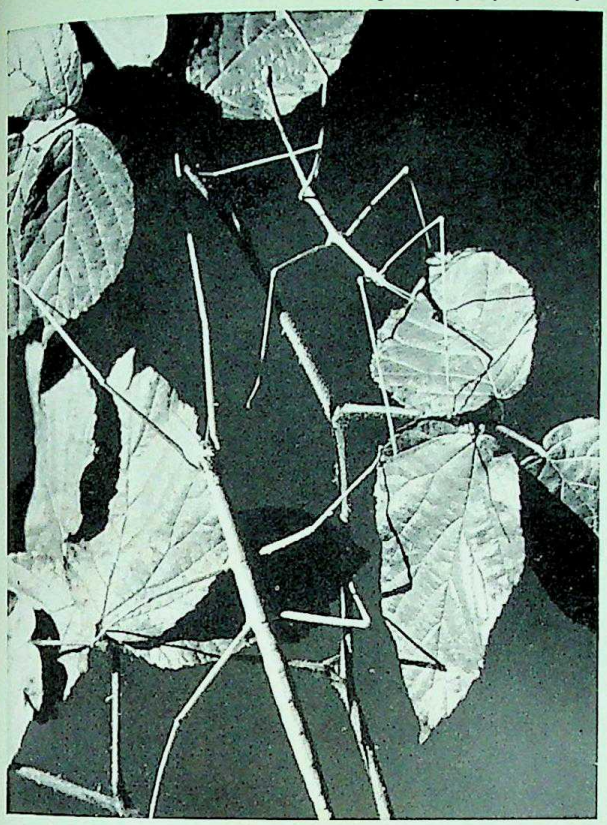

FIGURE 1 – Clitumnus extradentatus. *Male and female feeding on blackberry leaves. Note the sexual dimorphism of this apterous species. The male is much smaller and more fragile than the female.*

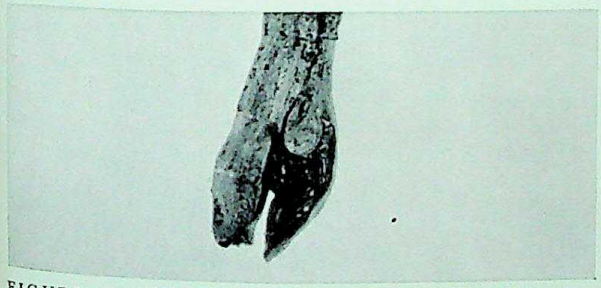

FIGURE 2 – *Abdominal extremity of an intersexed* Carausius morosus *(side view). The female subgenital plate is only partially developed on this side, enabling one to see the male copulatory organs.*

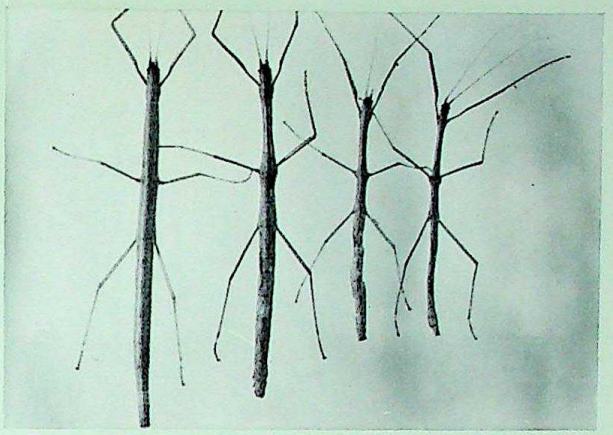

FIGURE 3 – Carausius morosus. *Series of four individuals exhibiting increasing masculinity. Left, female; right, 'male' (dorsal surface).*

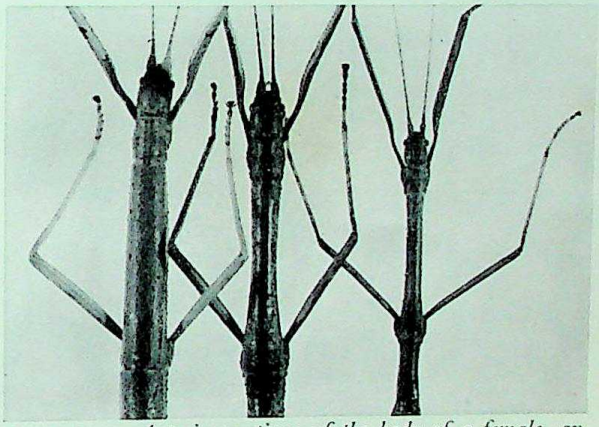

FIGURE 4 – *Anterior sections of the body of a female, an intersexed specimen, and a 'male' of* Carausius morosus. *The 'male' has a narrower thorax and a smooth cuticle.*

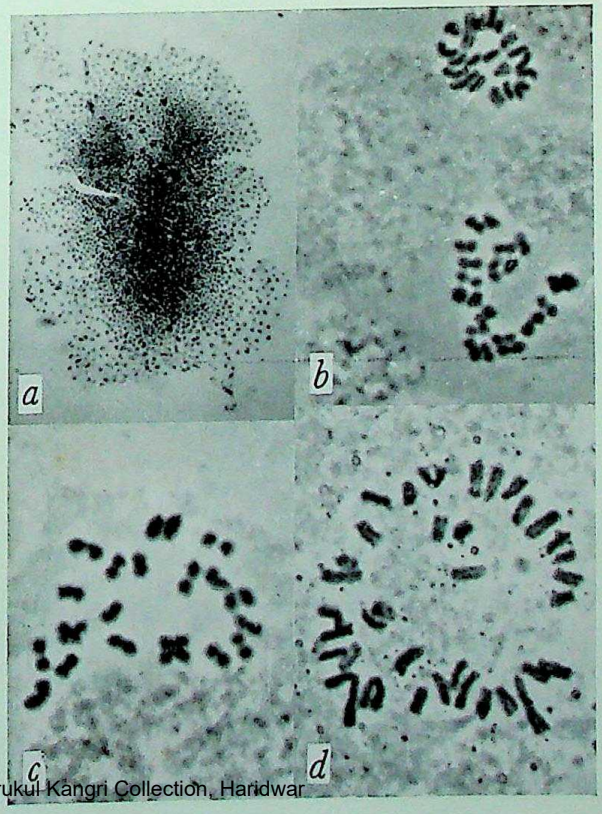

FIGURE 5 (*right*) – *Chromosome regulation during the facultative parthenogenesis of* Clitumnus extradentatus. (a) *Very young embryo comprising only one cell layer situated in the posterior and dorsal section of the egg* (× 100). (b) *Crushed embryo from an unfertilized egg; (above) haploid mitosis* ($n = 19$), *(below) blocked metaphase of which* (c) *is another example* (× 100). *The very condensed chromosomes are now attached only by their centromeres. No anaphase, and the cell subsequently develops towards the diploid state.* (d) *Diploid mitosis of a male* (37 *chromosomes*). *The only heterochromosome is the large V-shaped one.*

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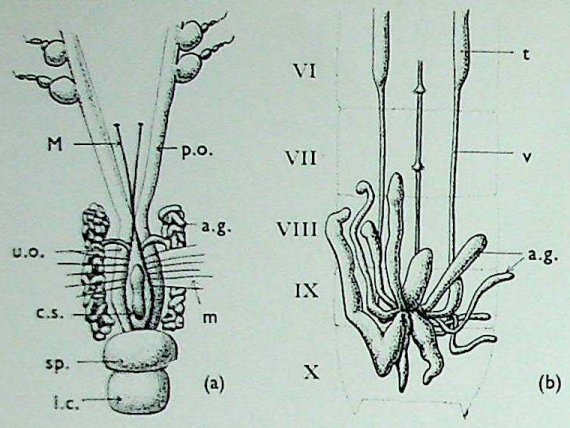

FIGURE 6 (a) – *Posterior section of the female genitalia in* Carausius morosus (*dissection through the dorsal surface*). *l.c., laying chamber; a.g., accessory glands; M, muscles of the copulative sac inserted behind the sixth sternite; m, dilator muscles of the copulative sac; u.o., unpaired oviduct; p.o., paired oviduct; c.s., copulative sac; spermatheca.* (b) *Dissection of the posterior section of the male genitalia of* Carausius morosus *through the dorsal surface. VI, VII, VIII, IX, X, abdominal segments; v, vas deferens; a.g., accessory glands; t, testicle.*

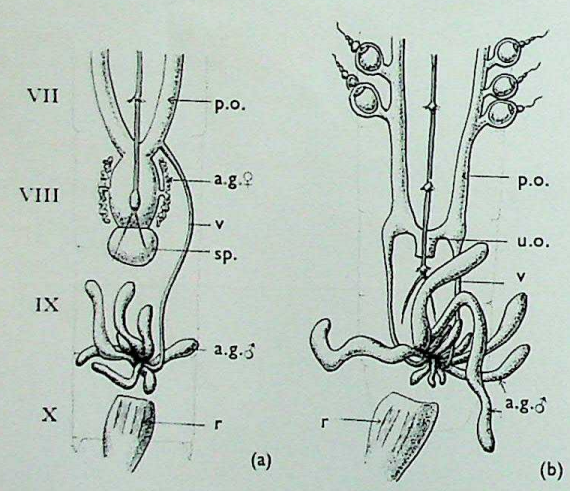

FIGURE 7 – *Posterior parts of the genitalia of two intersexed specimens of* Carausius morosus, *illustrating the coexistence of male and female ducts.* (a) *Female organs still fairly well developed and even exhibiting accessory glands. Vas deferens present on one side only.* (b) *Female organs smaller in the terminal section where there is only one common, non-differentiated oviduct. v, vas deferens; a.g., accessory glands; u.o., unpaired oviduct; p.o., paired oviduct; r, rectum; sp., spermatheca.* (*Cf. figure* 6)

present the two types of organ, male and female, side by side in a more or less rudimentary state. Thus, for example, intersexed insects often have a female subgenital plate, generally small and rather badly formed, and a more or less characteristic male subgenital plate (figure 2). The same is true of the genital ducts. After the gonad, which in this case is a typical ovary, intersexed individuals exhibit large channels (oviducts) joining together on the ventral surface at the level of the eighth abdominal segment to form a fairly well differentiated common oviduct. This receives the more or less rudimentary female accessory glands. But we also find spermiducts that end in the midst of a bundle of male accessory glands opening on the ventral surface between the ninth and tenth abdominal segments (figure 7). In intersexed specimens with little masculinity there is sometimes only a rudiment, of varying size, of these male accessory glands with no trace of spermiducts. Finally, certain weakly intersexed individuals remain completely female so far as their internal anatomy and external genital pieces are concerned. Only the characteristics of the cuticle and coloration of the ventral surface of the thorax give evidence of the mild treatment that has been applied. These weakly intersexed insects remain capable of oviposition and having normal progeny; strongly intersexed specimens produce eggs in their ovaries but are generally incapable of laying these because of inadequate differentiation of the terminal section of the female genital ducts.

These facts allow us to decide between gynandromorphism and intersexuality. The constant coexistence of the rudiments of the organs of both sexes, which may present any of the intermediate stages of development, shows that what is involved here is intersexuality. This is, moreover, not genetic, because the chromosome formula (64 chromosomes) remains unchanged in the animals treated and masculinity is increased by longer treatment.

The existence in Orthoptera embryos of the outlines of both male and female ducts for a period of embryonic life during the development of one of the two sexes has been known for some time. More detailed investigation has shown that *Carausius morosus* controls, incubated at the normal temperature of 23°C and destined to differentiate as females, exhibit the outlines of male ducts, derived from the ninth and tenth coelom sacs of the abdomen. These disappear, by pycnosis of the constituent cells, towards the 37th day of embryonic life. Incubating at 30°C at the beginning of embryonic development has the effect of maintaining this normally transitory male outline and allowing it to develop. At the time of treatment the gonad is not yet differentiated—its visible differentiation does not start until about the 50th day of the embryo's life. The secondary sexual characteristics are not differentiated until some

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months later, towards the end of the larval stage. It is interesting to consider the ties of a physiological nature that must be involved in harmonizing these various differentiations.

This is the first time the sex of an insect has been altered experimentally. T. Dobzhansky's work [7], it is true, showed the effect of temperature on the differentiation of intersexed genetic specimens of the vinegar fly (*Drosophila melanogaster*): here femininity was increased by rearing at an increased temperature; a similar phenomenon had been demonstrated by R. Goldschmidt and his co-workers in their classical work on *Lymantria dispar*. These, however, were intersexed individuals that remained sterile and intermediate between the two sexes whatever the breeding temperature; the experiments with *Carausius morosus* give a practically complete reversal of the sex of an animal of normal genetic constitution.

'Males' of *Carausius morosus* produced experimentally in this way generally exhibit abnormal spermatogenesis. The 64 chromosomes pair off as 32 bivalents, it is true, but in the majority of the cysts the metaphase of the first meiotic division is prolonged almost indefinitely, as is evident from the number of cysts found in this state. The bivalents separate one after the other in such a way that one occasionally finds univalents corresponding two by two in these metaphases. Nuclear division does not take place and there is no evidence of the second maturation division. The basis of spermatogenesis is present, however, and in most cases dissection reveals the presence of numerous abnormal giant spermatozoa with three or four flagella, probably a result of the non-division of a spermatogonium. In some individuals spermatogenesis is more normal, and spermatozoa of perfectly normal appearance can be found in the vas deferens. It was probably on the basis of such specimens that H. Pehani [11] described the spermatogenesis of *Carausius*.

CONCLUSION

The sexual biology of the Phasmids, therefore, appears to contain many aberrations. There are, depending on the species, two different reasons for a scarcity of males. It may be due to the dispersal of the species in small colonies that are facultatively parthenogenetic and produce few true genetic males. An example is *Bacillus rossii* in North Africa. Alternatively the 'males' are genetic females whose sex differentiation has been altered by climatic factors. These 'males' are nothing other than very masculine intersexed specimens that often have difficulties with spermatogenesis. This prevents them from fertilizing females, whose eggs are therefore obliged to develop parthenogenetically, a method that produces only females. Even if spermatogenesis is normal and fertilization takes place, it is difficult to prove, as all the spermatozoa in this case will have the same chromosome make-up as the ovum; they must, for example, all carry one X-chromosome. The issue of an improbable fertilization of this nature would be wholly female, as in the case of parthenogenetic egg development.

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The neuron and its glia—a biochemical and functional unit

HOLGER HYDÉN

Although neurochemistry is of relatively recent origin, it is now a flourishing field of research. The investigations are of interest not only because of their fundamental significance for the understanding of nervous function but because of the extreme delicacy of the biological and biochemical manipulations. The experimental evidence indicates a very close relationship between the neuron and its glia, the latter apparently constituting an effective feedback mechanism co-ordinating the electrical and biochemical processes in the neuron.

Neurons—supported by the glia—are the structural units of the nervous system (figure 2), and their combined activity controls the complex activities of living animals. The neuroglia was first described in 1846, by Virchow. The name—from γλία, meaning glue—gives a good indication of its consistency, and reflects the idea that the glia constituted a supporting substance. In man the neurons are numbered in hundreds of millions; Sir Charles Sherrington [3] likened the brain to an enchanted loom where millions of flashing shuttles weave a dissolving pattern, always significant but never lasting. Although the fundamental importance of neurons in nervous activity has long been recognized, it is only comparatively recently that their function has begun to be interpreted in physical and chemical terms.

The most striking manifestation of activity in a neuron is a change in electrical potential. Changes in electrical potential are fairly easily measured, and electrophysiologists have contributed much to our knowledge of the nervous system. Primarily because of difficulties of technique, but also because its possibilities were not sufficiently widely recognized, the chemical approach was made later. Today, however, neurochemistry is a flourishing field. Although we have a long way to go before we can have anything like a complete picture of the biochemistry of intact neurons and glia, it is now possible to correlate some aspects of molecular change with functional activity. In electrophysiology, the present tendency is to concentrate on phenomena in single neurons or parts of neurons. In neurochemistry the emphasis is on characterizing physiologically active substances, locating them in the cell, and investigating their metabolism.

The biochemistry of the neuron presents two major practical difficulties. Firstly, there is the difficulty of isolating material without serious damage. Secondly, there is the problem of analysing cell constituents present only in 10^{-12} g quantities.

SAMPLING OF MATERIAL

The collection of fresh clean samples of neurons or glia is difficult. Both types of cells have extensively branching processes that are intimately interwoven. Furthermore, electron-microscopic studies have shown that the glia consist mainly of thin delicately folded membranes, which entirely cover the neurons except for the synaptic knobs and capillaries. For these mechanical reasons, separation of glia and neurons by differential centrifugation has so far proved unsatisfactory. Figure 2 indicates the morphological relationship between neuron, glia, and capillaries.

Separation of the two types of cell is essential, however, because the biochemical changes that follow stimulation may go in opposite directions in neuron and glia. Unless each is analysed separately, a clear picture cannot be obtained.

For certain types of analyses, micromanipulators can be used for the dissection of cells from precipitated and embedded tissue. Such a procedure is not suitable, however, for fresh tissue,

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Was born in Stockholm in 1917 and educated at the Karolinska Institute, where he studied the distribution and concentration of nucleic acids in the nervous system with microspectrographic methods. He became assistant professor in 1943 and was elected to a research fellowship at the Nobel Institute for Cell Research in 1945. He became head of the Department of Histology at the University of Gothenburg in 1949. The recently erected research building is also an Institute of Neurobiology, of which he is director. Among his many papers on the subject is a monograph 'The Neuron' in Vol. IV of *The Cell* (ed. by J. Brachet and A. E. Mirsky, 1960).

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when neurons and glia must be sampled rapidly. In our laboratory at Gothenburg we isolate individual nerve cells and glia by free-hand dissection, using a binocular microscope of about one hundred times magnification [4]. The dissecting instruments used are made from stainless steel threads, 15 μ in diameter; the thread can simply be given a point, or, by cold forging, a spatula or hook can be formed. The instrument is inserted below a nerve cell at the surface of a thick nerve-tissue section immersed in an isotonic solution. The cell, together with surrounding glia, is lifted up and transferred to clean solution. By gentle manipulation under the microscope, the nerve cell—including the first part of the dendrites, about 100–200 μ long—is freed from glia membranes; figure 4 shows a nerve cell isolated in this way. The glia cells originally surrounding the nerve cells tend to adhere to each other; the trimmed-down collection of glia has about the same volume as the nerve cell. The glia and their nerve cell so prepared have the same dry weight, as has been shown by X-ray microradiography [5]. Figure 5 shows two separated nerve cells with their isolated glia [5].

Nerve cells and glia isolated in this way are not vitally damaged, as is shown by the fact that it has been possible to culture them in this laboratory for periods up to three months [6].

This method of isolation has now been extended to isolate fresh membranes from the nerve cells [7]. In this technique about ten nerve cells are rapidly isolated and placed on a glass slide in sucrose solution. A longitudinal cut is made—with a stainless-steel knife 20–30 μ long—along one of the dendrites of the nerve (figure 7). The flaps of the cell are folded back, and the cell content is removed partly mechanically and partly by directing a jet of solution on to it from a micropipette. Figure 6 (*a*) shows a nerve cell photographed at 2600 Å, and figure 6(*b*) the same cell under the phase-contrast microscope; the isolated cell membrane is seen in figure 6(*c*). The nerve cells illustrated in figures 2, and 4–7 are the big cells known as Deiters' nerve cells, and it is on these that most of the analytical work described below has been carried out. The small, dense particles seen scattered over the membrane are synaptic knobs. Membranes isolated in this way were measured interferometrically and found to be 2500–2900 Å thick [7]. These membranes consist of the 180 Å thick double-layered structure seen with the electron microscope, plus a thin, largely structureless, layer of the cell body. Capillaries of fresh nervous tissue also can be rapidly isolated.

METHODS OF ANALYSIS

Analytical methods must take account of the fact that a medium-size nerve cell weighs only 10^{-10} g. Methods capable of estimating gamma quantities, which in many fields would be considered very sensitive, would thus have no significance in studying individual nerve cells. Ultramicrotechniques developed for neurochemistry make it possible to detect quantities in the 10^{-9} to 10^{-11} g range. These techniques will be described in relation to the estimation in single cells of RNA and its component bases; total protein and dry weight; and enzymes.

Neurons have a remarkable capacity for producing RNA, and this is linked with functional activities of the nervous tissue. Medium and large neurons of mammals contain from 40 to about 2000 μμg of RNA [8]. In general, RNA seems to be produced in the nuclei of cells, and the neuron seems to be no exception. The site of RNA synthesis is partly in chromosomes, but the greater part of it is probably transported to the nucleolus and thence to the cytoplasm [9]. The mechanism of transportation, as well as the question of independent production of RNA in the cytoplasm, is still under discussion [10].

The role of RNA in protein synthesis is now beginning to be understood in some detail. The DNA of the chromosomes is the carrier of genetic activities, generally regarded as encoded in the sequence of purine and pyrimidine bases. By a base-pairing mechanism more genetic nucleic acid can be formed. A small fraction of the RNA, of low molecular weight, has been found to have the highest turnover. There is evidence that in bacteria this RNA fraction can act as a primer for the synthesis of protein in the cytoplasm; it has therefore been called 'messenger' or 'tape' RNA.

In the synthesis of cytoplasmic proteins, each amino acid—linked to a separate RNA chain—is activated by an enzyme system, leading to formation of aminoacyl RNA [11]; each enzyme seems to be specific for the formation of only one aminoacyl derivative. It is believed that the soluble RNA acceptor, after accepting the activated amino acid, is transferred to the highly polymerized RNA of the ribosomes in the cytoplasm. Through a base-pairing mechanism the amino acids reach their specific positions; and thus polypeptides and proteins—including enzymes used for the synthesis of other substances—are formed.

Cytoplasmic protein synthesis takes place in the ribosomes; these are part of the so-called endoplasmic reticulum, and are small particles situated

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on thin membranes that enclose submicroscopic, flat sacs.

The fundamental importance of RNA in cellular metabolism is the reason why it has been worth while to develop extremely sensitive methods for its estimation.

DETERMINATION OF RNA

RNA in μμg amounts can be determined as follows [12, 13]. Isolated nerve cells, or a weight of glial cells equal to that of the nerve cell, are precipitated and a 0·5 mμl microdrop of a buffered solution of ribonuclease is added (figure 1); the operation is carried out in a chamber filled with paraffin oil. The sample is then extracted three times, for a total of sixty minutes, and the extracts, containing depolymerized RNA, are transferred to a clean quartz cover slip. The buffer solution, which contains ammonium bicarbonate-acetate, rapidly evaporates, leaving the depolymerized RNA as a solid residue, which is dissolved in a microdrop of glycerol-containing buffer. This microdrop, 100–200 μ in diameter, is optically homogeneous and contains all the RNA present in the original sample. It is photographed at 2600 Å, together with a reference system consisting of a rotating disc with logarithmically cut steps. The extinction at this wavelength is evaluated photometrically. By this method, amounts of RNA down to 20 μμg can be determined with an accuracy of ± 5 per cent.

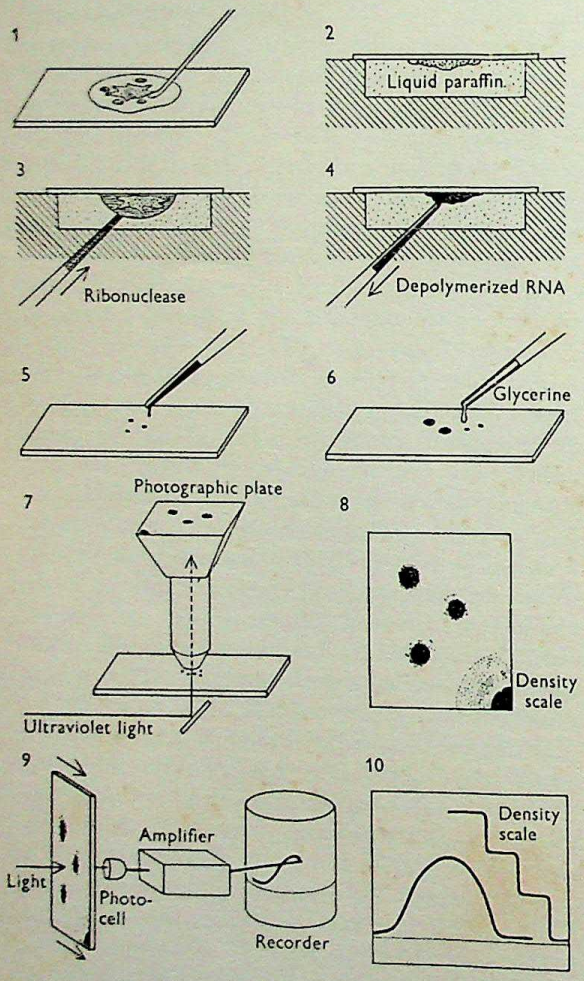

FIGURE 1 – *Determination of RNA. Isolated nerve cells are placed on the lower side of a glass slide and covered with paraffin oil (1, 2). A buffered ribonuclease solution depolymerizes RNA, which goes into solution (4). The solvent evaporates, and the RNA is dissolved in a buffer containing glycerol (6). The microdrops are photographed at 2600 Å, together with a reference system (7, 8). The evaluation is made by a microphotometer (9), the RNA content of each drop being obtained from the extinction curves (10).*

Table 1 shows that different types of cells have widely differing RNA contents. Thus a small neuron may contain 45 μμg, while the large vestibular nerve cells in Deiters' nucleus may contain more than 1500 μμg, as the nuclei of the latter cells contain 55 μμg of RNA. The neuronal glia of Deiters' nerve cells contain 130 μμg of RNA, that is, about one-tenth of the quantity contained in the nerve cell itself. Apart from total content, we also have some information about the distribution of RNA within individual nerve cells. Approximately 10 per cent of the nerve-cell RNA is contained in the dendrites. There is as yet no information on the RNA content of the axons of mammalian neurons, but recently J. -E. Edström, working at our laboratory with big neurons from fishes, has found the axon to contain around 6000 μμg of RNA and the perikaryon 4000 μμg [14]. No other cells in the organism are known to contain RNA in such amounts.

TABLE 1

RNA content in different types of nerve cells

Type of nerve cell	RNA (μμg/cell)
Retinal gangl. cells, rabbit	45
Nerve cells of the supraopt. nucl., rabbit	70
Spinal gangl. cells, rabbit	1070
Hypoglossal cells, rabbit	200
Ant. horn cells, rabbit	530
Ant. horn cells, man, 40–50 years ..	670
Ant. horn cells, man, 60–70 years ..	540
Deiters' giant cells, rabbit	1550
Deiters' nucleus medium-size cells ..	885

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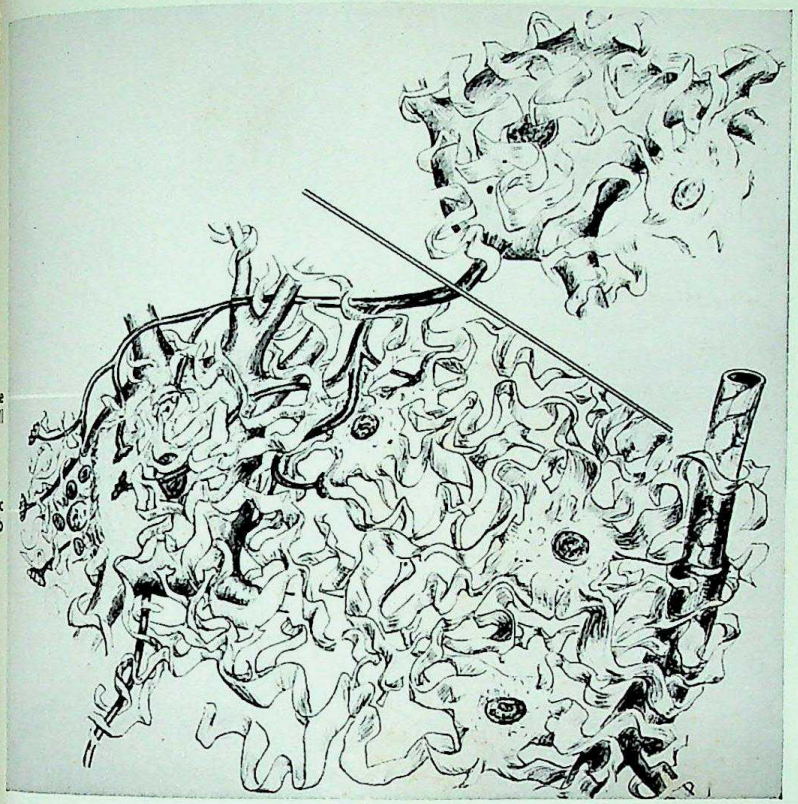

FIGURE 2 – *Drawing showing relationship between the neuron and the glia. The large cells on the left are nerve cells. These, with their processes, are called neurons. To the right is a capillary; both neuron and capillary are covered by thin glial membranes.*

FIGURE 3 – *Enucleated nerve cell; the nucleus is seen above.* (2600 Å; ×200)

FIGURE 4 – *Freshly isolated nerve cell from the lateral vestibular nucleus of a rabbit.* (*Phase contrast*, ×500)

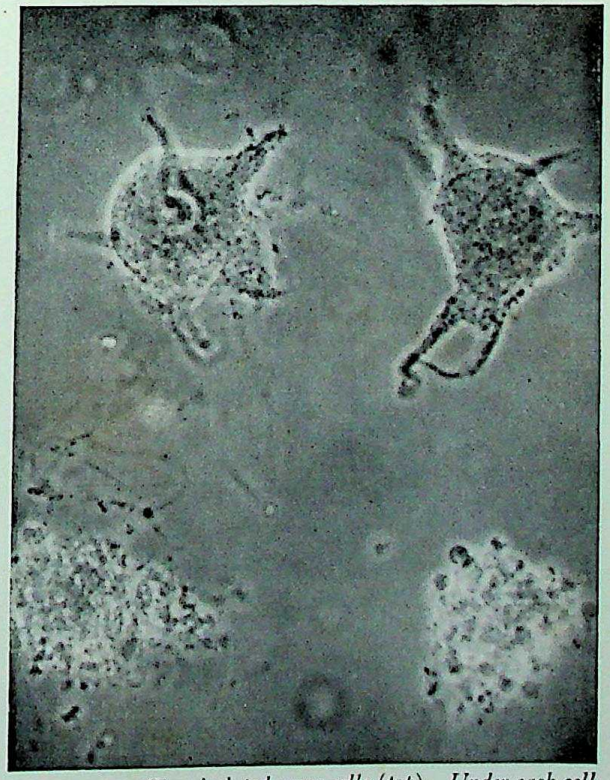

FIGURE 5 – *Two isolated nerve cells (top). Under each cell is the glia that had originally surrounded it. The collection of glia is the same as the nerve cell which it surrounded.* (×230)

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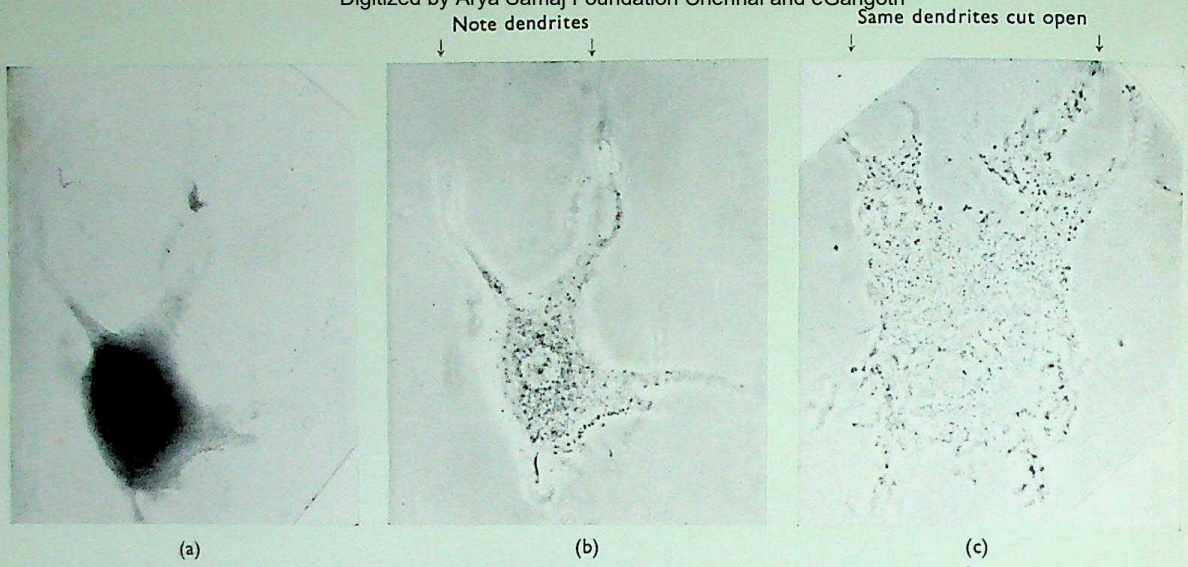

FIGURE 6 – (a) *Fresh Deiters' nerve cell, treated as in figure 7, photographed in radiation of 2600 Å wavelength.* (b) *The same, photographed under phase contrast.* (c) *The membrane of the nerve cell seen in* (b). (×200)

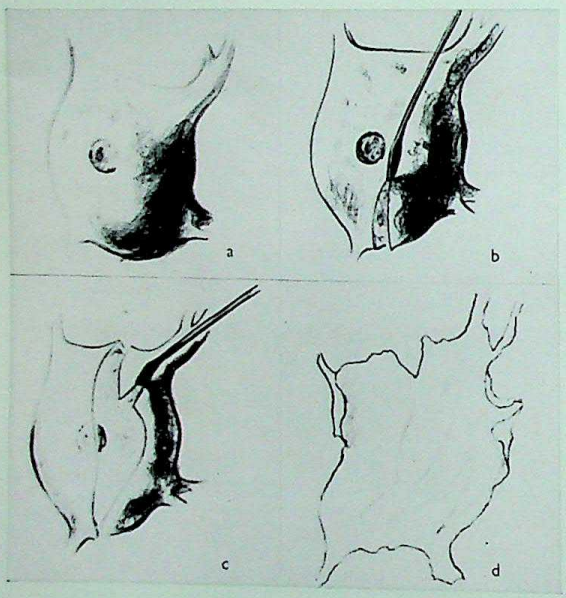

FIGURE 7 – *Technique for microsurgical preparation of nerve-cell membranes.* (a) *Isolated nerve cell.* (b) *A cut is made along the cell.* (c) *The cell is further opened by a second cut; the flaps are folded back, and the cell content is removed.* (d) *The isolated cell membrane, about 2500 Å thick.*

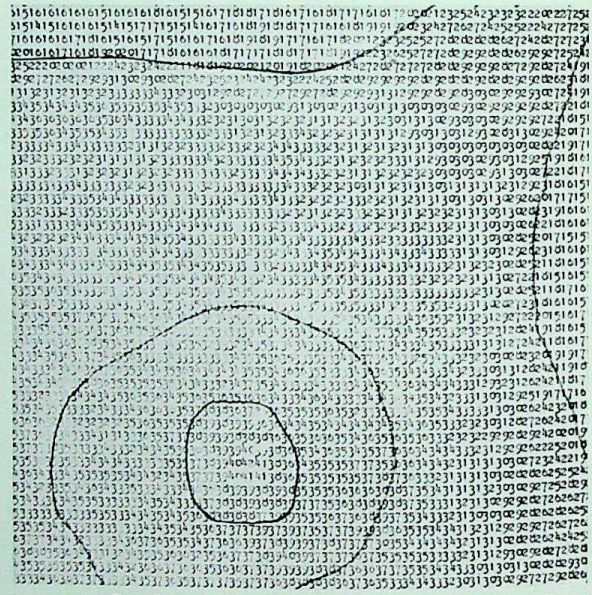

FIGURE 8 – *Pattern in figures given by the cell analyser described in figure 10, showing the distribution of proteins within a nerve cell.*

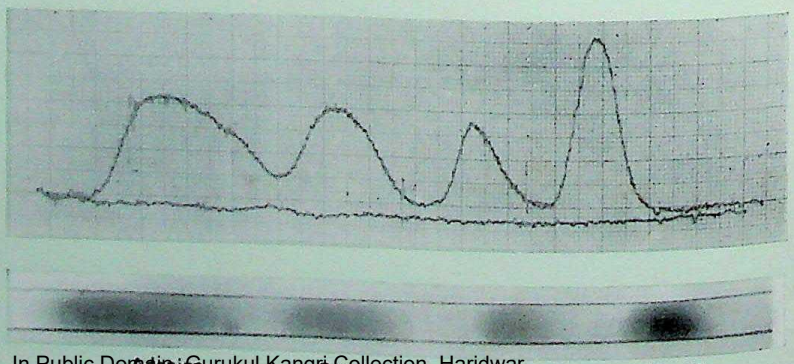

FIGURE 9 – *Microelectrophoretic separation of RNA from a nerve cell; the photometric recording is seen above.*

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COMPOSITION OF RNA OF THE CENTRAL NERVOUS SYSTEM

What is the composition of the RNA of the central nervous system? Does it differ from the RNA of other cells? Do the nerve cells and the glia contain the same type of RNA, in terms of constituent bases? These questions can now be answered by Edström's microelectrophoretic method.

The RNA is extracted from the sample in the way described above. After drying, it is hydrolysed in a micropipette with 4N HCl at 100°C for thirty minutes (figure 11). The purines and pyrimidines are then determined electrophoretically, using a microscopic cellulose fibre (diameter 20 μ) treated with a buffer of high viscosity.

The hydrolysate is placed on the fibre, which is then transferred to a constant-humidity chamber. A potential gradient of 2000–3000 volts per cm is applied for ten minutes, after which the separation will be complete. The fibre is photographed at 2600 Å, and the variation of optical density along it is recorded with a microphotometer (figure 9). From the recorded curves the purine and pyrimidine bases can be estimated as percentages of the total. For 500 μμg samples of RNA the determinations show a coefficient of variation of ± 5 per cent.

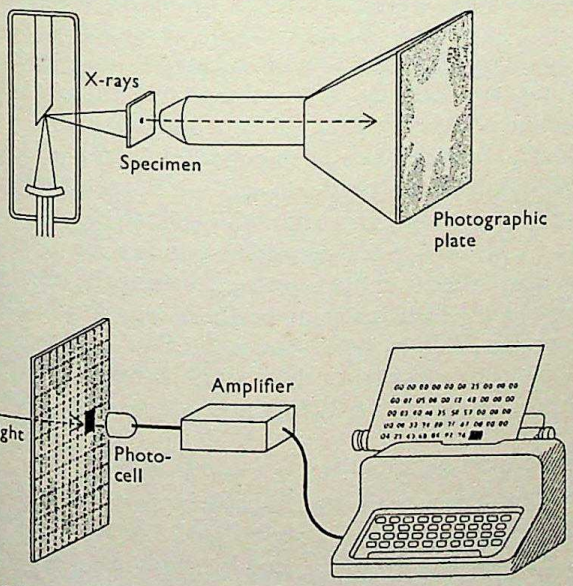

FIGURE 10 – *Quantitative X-ray microradiography for the determination of dry weight, proteins, and lipids in nerve-tissue components. An isolated, dry nerve cell is photographed using radiation of 10Å wavelength. A scanning analyser which makes an automatic record divides the X-ray microradiograph into 12 000 areas, each with an area of about 3 μ². The computer translates density values into the quantity of substance per area. The writing unit expresses the result in two figures, ranging from 00 to 99 (figure 8).*

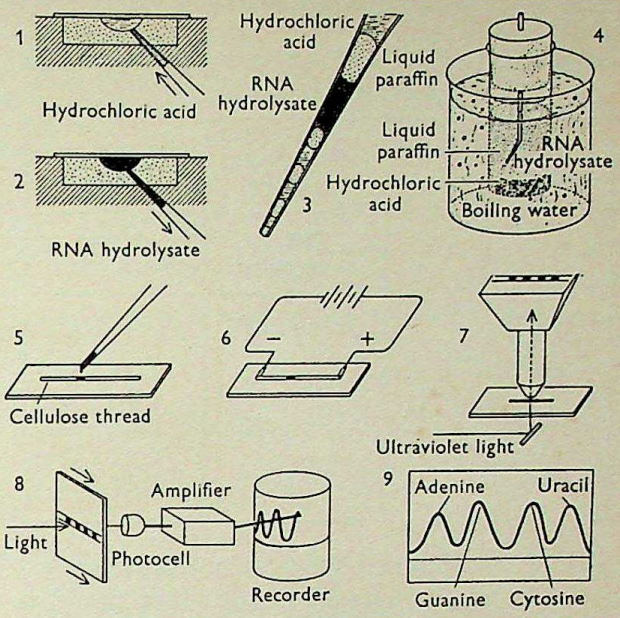

FIGURE 11 – *Determination of the base composition of RNA. Depolymerized RNA is hydrolysed by hydrochloric acid (1, 2) in a water-bath (4), and the hydrolysate is taken up in a micropipette (3). The hydrolysate is placed on a fine cellulose thread treated with a buffer of high viscosity (5), and an electric field of 2000–3000 volt/cm is applied to effect separation (6). The electrophoretic pattern is photographed at 2600 Å (7). Extinction curves of adenine, guanine, cytosine, and uracil are obtained by microphotometry, and from them the molar proportions are determined (8, 9).*

Table II shows that nervous-tissue RNA shows characteristically high values of the bases guanine and cytosine [15]; it is thus a markedly 'cytoplasmic' type of RNA. The RNA of the nerve-cell nucleus shows higher contents of adenine and uracil (table III), in this respect more like DNA [16]. By analogy with findings from spider oocytes [17], the nuclear RNA from nerve cells can be described as being of a 'nucleolar' type. Figure 3 shows a nerve cell from which the nucleus, which can be seen above the cell surface, has been removed. The cell was photographed at 2600 Å, hence the high absorption by purines and pyrimidines. As can be seen from table II, the composition of the nerve-cell RNA differs from that of the

TABLE II

Relative molar proportions of bases in nervous-tissue RNA

	Nerve cell	Glia
Adenine	19·7	20·8
Guanine	33·5	28·8
Cytosine	28·8	31·8
Uracil	18·0	18·6

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TABLE III

Composition of RNA in Deiters' nerve cells, control nerve cells, and their nuclei

	Nerve cell cytoplasm	Nerve cell nuclei
	Molar proportions	Molar proportions
Adenine	19·7 ± 0·37	21·3 ± 0·42
Guanine	33·5 ± 0·39	26·6 ± 0·27
Cytosine	28·8 ± 0·36	30·8 ± 0·38
Uracil	18·0 ± 0·18	21·3 ± 0·43

glia. The neuronal RNA contains relatively more guanine and less cytosine than does the glial RNA.

X-RAY MICRORADIOGRAPHY

In the 8–12 Å region, X-ray absorption is proportional to the amount of carbon, nitrogen, and oxygen, and use has been made of this for analytical purposes. The original method [18] has been adapted to give sufficiently accurate values of the total amount of dry organic material in the nerve-tissue samples, and it is further possible to estimate how this is divided between a protein and a free-lipid fraction [19].

The fresh, dry nerve cells or glia are placed on aluminium foil 0·2 μ thick and irradiated on a high-resolution photographic film; a reference system consisting of a step-wedge made from nitrocellulose foils having the same wavelength dependence as that of the biological material is exposed together with the cells at 8–12 Å. The varying densities in the X-ray picture are recorded and translated into values of the amount of C, N, and O by an automatic scanning and computing analyser [20, 21] (figure 10). The scanning unit divides the sample into 12 000 areas, ranging in size from 0·75 to 6 μ^2, and measures the density of each; the computer can analyse 50 such areas per second. The final density value for each area is obtained by letting the signals pass an electronic function transformer, which converts nonlinear curves to linear ones on which every point is proportional to the organic mass. The analyser gives the C, N, and O value for each small area as a two-digit number, ranging from 00 to 99. The instrument can give the required information in either of two forms. The writing unit can print a pattern of figures showing the distribution of particular substances, for example proteins, within the cell (figure 8). Alternatively, it can give the integrated sum directly.

Table IV gives some results obtained with Deiters' nerve cells from the lateral vestibular nucleus of the rabbit. Such a cell has a weight of about 20 000 μμg, 16 000 μμg of which constitute proteins and the rest lipids extractable with chloroform. Typically, the neuronal glia contain 70 per cent lipids.

MEASUREMENT OF ENZYME ACTIVITY

The microtechnique described here was originally developed by E. Zeuthen [22] and has proved to be of great value, because we needed a method by which fresh glia and nerve cells could be analysed separately. One nerve cell is sucked into a small Cartesian diver having a weight of 0·2 mg and a gas volume of 0·01–0·02 μl. The cell is placed in a special substrate and the diver is so balanced that it remains suspended; if the weight is changed, the diver will change its position vertically until a new equilibrium is reached. When respiratory enzymes are being determined, oxygen is consumed and the air bubble in the chamber gets smaller. As a result, liquid is drawn in through the open part of the diver to replace the gas consumed, and the diver therefore sinks slightly. From the rate of sinking the oxygen consumption can be determined and the enzyme activity expressed as $\mu l\ O_2 \times 10^{-4}$ per cell per hour. Results have been reported for the activity of cytochrome oxidase, the succinate-oxidizing enzyme system, and anaerobic glycolysis.

O. H. Lowry and his collaborators have utilized measurements of fluorescence to study the enzyme activities of isolated nerve cells or very small tissue samples. They have also refined a method by which substances of biological interest can be estimated with a pyridine nucleotide system [23].

THE NEURON AS RNA-PRODUCER

Both structurally and biochemically, the neuron has the character of a specialized enormous gland cell. As has already been pointed out, the big neurons are outstanding as producers of RNA. The only cells that seem to be comparable are those in the exocrine part of the pancreas. The neuronal content of RNA is not constant, however, throughout life. In man there is a typical, slow change [8]. The amount of RNA per anterior-horn cell in the spinal cord increases significantly from birth to around 40 years of age (figure 12).

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TABLE IV

Composition of a Deiters' nerve cell

Component	Nucleolus	Nucleoplasm	Cytoplasm	Dendrites	Nuclear cell membrane
Total weight/volume	100 μμg	800 μμg	20 800 μμg	—	—
Dry weight/volume	0·38 μμg/μ³	0·21 μμg/μ³	0·23 μμg/μ³	0·20–0·10 μμg/μ³	—
RNA total	—	50 μμg	1 500 μμg	~ 100 μμg	Concentric proteins
RNA conc. weight/volume ..	1·5%	~0·5%	1·2–1·5%	—	—
RNA conc. (% of dry protein) ..	—	—	7%	—	Radial lipids (cerebrosides)
Proteins (total)	—	650 μμg	16 600 μμg	—	—
Proteins (conc., weight/volume) ..	—	16%	20%	—	—
Proteins (weight/cell weight) ..	—	—	75%	—	—
Lipid fraction	—	200 μμg	4 200 μμg	—	—
Lipids (conc., weight/volume) ..	—	5%	5%	—	—

From 40 to 55 or 60 years of age the amount of RNA per cell seems to reach a plateau, but after 60 it falls. The graph is based on 460 RNA analyses of material taken from cases of sudden death, such as motor-car accidents. Similar changes have been found in nerve cells from experimental animals. For this reason great care has to be taken in experiments always to use controls of the same age and nutritional condition.

What is the reason for this increase during the first part of life? It may be directly associated with the total amount of sensory stimulation of the cells.

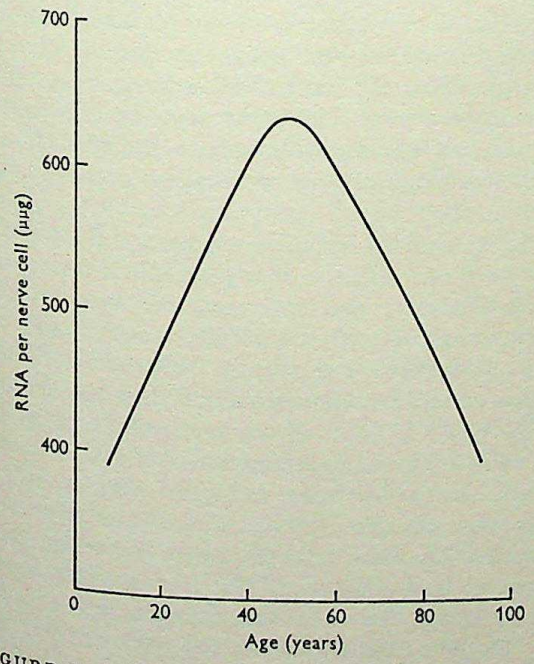

FIGURE 12 – *Amount of RNA in human motor-nerve cells of man at ages from 3 to 90.*

If a neuronal path, including its receptors, is deprived of the normal amount of stimulation during the critical period immediately after birth, the neurons will not develop biochemically or functionally. For example, if rabbits are born and reared in complete darkness, the retinal ganglion cells remain low in RNA and protein but have a higher water content than control cells [1]. It has also been shown that individuals deprived of light stimuli during a period early in life never develop functionally, or do so only partially [2]. On the other hand, if a motor or sensory part of the central nervous system is stimulated within physiological limits, each nerve cell affected will increase its content of RNA, proteins, and lipids [24, 25]. The increased production begins immediately the level of stimulation is increased. When the stimulation is decreased, the values return to starting level within a matter of hours. Such changes have been recorded in many cases, for example in the Deiters' nerve cells [5]. In a recent series of experiments, young rats were trained to balance on a thin wire one metre long and inclined at an angle of 45°. After being trained for one hour daily for four days, the average increase of RNA in the nerve cells was found to be 12 per cent, which was significant; even the composition of the base ratio was altered.

RELATIONSHIP BETWEEN THE NEURON AND ITS GLIA

The glia immediately surrounding the body of the nerve cell is called neuronal glia. As has already been pointed out, the glia contains relatively much more lipid than the neurons but only about one-tenth of the amount of RNA. Another

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difference is that while the glia oxidizes mainly succinate and pyruvate, the neuron utilizes mainly glutamate [26]. The biochemical data indicate that the glia provide a maintenance system for the neuron, the two together effecting a well-balanced metabolic symbiosis.

Although a vast amount of knowledge has been collected about the neuron, information about the glia is scarce. The electro-physiological methods that have proved so powerful in analysing the function of neurons have contributed little to our knowledge of the properties of glia. The neuronal glia show slow contractile movements, which can be elicited by electrical stimulation, and last for about five minutes [27]. The glia have a resting potential of 50 mV, and on stimulation give potential changes that are about 1000 times slower than those generated by neurons [28]. The neuroglia outnumber the neurons by a factor of about ten. The total volume of both cell types is approximately the same. The neuron does not ordinarily undergo mitosis; we are born and die with the same neurons, apart from those that degenerate with increasing age. The glia, on the other hand, can multiply by mitosis.

Biologically important results have been obtained by cell culture, in which it has been found that only neurons surrounded by glia will grow for any length of time. In cultures, glial cells are observed to climb along the neurons like spiders, demonstrating a pronounced affinity for them [29]. Pinocytosis occurs in glia; this term was originally coined to designate the capacity of cells to take up small drops of fluid through the membrane, but it is now clear that pinocytosis consists primarily in the absorption and uptake of molecules through cell membranes.

There seems to be no 'extracellular' space in the central nervous system. The space between the neurons is completely filled by the glia, the delicately folded membranes of which, separated by about 200 Å, intertwine and cover every part of the neuron except the synaptic knobs. It is generally believed that all substances from the blood that are taken up by the neuron must pass through the glia. The glia may constitute a blood–brain barrier, letting certain substances pass into the nervous tissue but prohibiting others, thus maintaining the constant environment that the central nervous system enjoys.

How do the neuronal glia react to stimulation? To gain some information on this, the vestibular organ in rabbits was stimulated for 25 minutes daily for from one to seven days. The animal was placed, with the head peripherally, in a tight-fitting box on a circular disk. Stimulation was carried out by rotation through 120° horizontally and 30° vertically; the frequency was 20–30 turns per minute [5].

The biochemical changes in the glia were found to be the opposite of those in the neuron. The RNA and proteins increased in the neurons but decreased in the glia. The respiratory enzyme activities rose by 50–70 per cent in the neurons and fell about 70 per cent, as measured by cytochrome oxidase activity, in the glia. On the other hand, the anaerobic glycolysis—measured in terms of carbon dioxide production—fell in the nerve cell but rose in the glia. In the two types of cells a phenomenon with the same effect as the Pasteur effect (inhibition of carbohydrate metabolism by oxygen) could thus be seen, if they were looked upon as an interlinked system of two units. If the glia changes are considered, the increase in anaerobic glycolysis when the respiratory enzyme activities fell are indicative of a Crabtree effect [30], that is, suppression of mitochondrial oxidation by glucose.

It is interesting that the anaerobic glycolysis increased in the glia when the cytochrome oxidase activity rose in the nerve cell. The efficiency of energy utilization is around 55 per cent when the glucose is oxidized via the Krebs cycle and respiratory chain; it is only about 3 per cent when the cell uses anaerobic glycolysis. The glia are therefore the more modest in energy requirements, and leave the neuron to utilize the easily available energy when the level of stimulation rises. This was contrary to the effect seen in the control (unstimulated) animals, where the glia showed twice as much respiratory enzyme activity as the nerve cell.

In spite of the opposite response in the glia and in the nerve cell, the changes did not correspond quantitatively, and it cannot be assumed that enzymes are directly transferred from glia to nerve cell. Nevertheless, the results indicate that the two types of cell are energetically linked together.

Next, a kinetic study was made of the activity of the succinate-oxidizing enzyme-complex in living vestibular nerve cells and glia after increasing duration of stimulation of the type described above. Enzyme activities were expressed on a dry-weight basis [31]. The temperature dependence of the activities was studied at 37°, 32°, and 27°C. The results (figure 13) show a clear difference between the neuron and its glia. The nerve cell reacted by greatly increasing the capacity of

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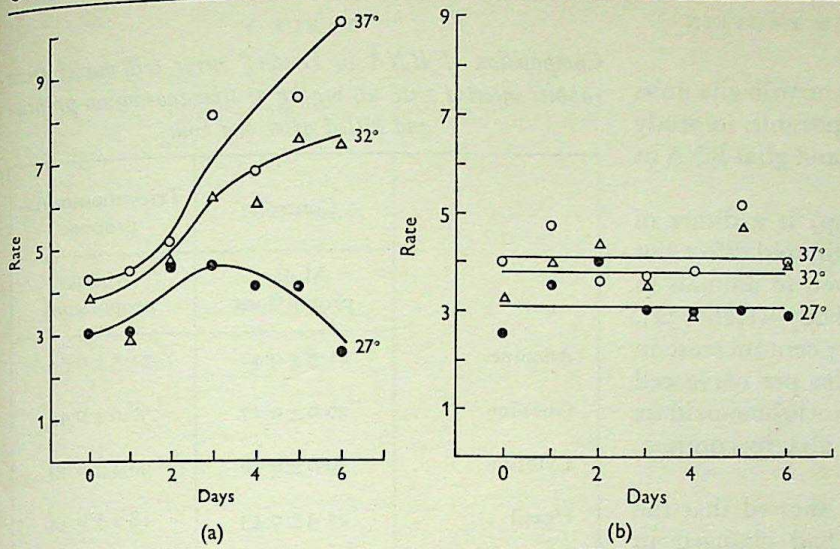

FIGURE 13 – *Changes in the activity of the succinate-oxidizing enzyme system as a function of duration of stimulation at* 37°, 32°, 27°*C in* (a) *Deiters' nerve cells and* (b) *their glia cells.*

the electron-transporting system, reflecting an increased consumption of energy as a function of the stimulation. The glia did not change in this respect; the rate of the enzyme reaction had no relation to the duration of the stimulation. When animals were subjected to decreased oxygen tension (8 per cent O_2 for 12 hours) neurons showed greatly increased oxidative enzyme activity; the glia did not, but showed increased capacity for anaerobic glycolysis.

The kinetic studies, together with the other observations, show that the nerve cell has higher energy requirements than the glia, and that the two are linked energetically. They confirm that when the functional demand increases, the neuron has first call on the available energy supply.

The energy requirements of nerve-tissue structures are intimately connected with the production of proteins. The activity of adenosinetriphosphatase (ATP-ase) and the characteristics of that enzyme complex were therefore studied.

The ATP-level was found to be high in nerve cells but low in the glia [7]. ATP-ase releases energy from ATP, and the amount present indicates the energy-utilizing ability of the cells. In view of the functions and rapidity of ion transport in the central nervous system, the ion requirements of ATP-ase were also characterized. For this, a radioactive method was worked out, having a sensitivity of 5×10^{-12} g of ATP hydrolysed per sample per hour.

The neuronal glia gave a rate of 1·7 μμmole per hour, indicating an 'ecto' ATP-ase. The value for the capillary glia was 50 per cent lower. On the outside of the intact nerve cell there was no ATP-ase activity. The glia ATP-ase was not activated by Na^+ but by K^+. The results suggested that the very active ATP-ase of the glia serves as a K^+- regulator in the extra-neuronal space. Since the thin glial membranes lie flush on the cell surface, the ATP-ase could increase the ionic potential across the nerve-cell membrane. On homogenization a nerve cell showed 1·0 μμmole of ATP hydrolysed per hour; the maximum hydrolysis was at pH 7·4, contrasting with the glia, for which it was at pH 8·0. The isolated nerve-cell membrane was found to possess 25 per cent of the cell ATP-ase activity that was not stimulated by Na^+. With Na^+ and K^+ together, the activation was higher than that found in media containing sucrose alone or Na^+. This 'endo' ATP-ase has the characteristics of a transporter ATP-ase. The activity was completely inhibited by ouabain. It could supply energy to the ion-pump of the nerve-cell membrane, and also serve to transport any substance across the membrane. From the aspect of the ATP-ase complex there are thus differences between the glia and its neuron, and the results give additional evidence of a glia-neuron unit functioning in a metabolic symbiosis.

Substrate must be transported from the capillaries to the neuron through the layers of glial membranes. The 'ecto' ATP-ase of the glia may serve to effect this transport. The ATP-ase would also have a second function intimately connected with the production of the neuronal potentials. Rapid movements of Na^+ and K^+ through nerve cell and axon membranes occur at the production of the nerve impulses. When the nerve impulse passes a given point, Na^+ ions flow into the cell; an instant later, in the wake of the impulse, K^+ ions briefly flow out. In the resting state, an energy-requiring mechanism continuously pumps Na^+ out of the cell and K^+ into it, thus creating the conditions necessary for the propagation of the nerve impulse. Our ATP-ase data indicate a balanced system for active transport, the extra-neuronal part of which furnished substrates and regulates the ionic potential across the nerve cell membrane. The 'endo' ATP-ase of the membrane indicates that Na^+ and K^+ transport is coupled.

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INDUCED RNA CHANGES WHEN PROTEIN PRODUCTION IS INCREASED

Using chemical substances and neuron glia units as model systems, it has been possible to study interrelated changes of neuronal and glial RNA at molecular levels.

Tricyano-amino-propene (triap) is a dimer of malononitrile which has an antithyroid effect but no observable toxic effects if given to animals in doses of milligrams per kilogram body-weight [32]. In one hour triap caused a 25 per cent increase in the quantity of RNA and proteins per nerve cell [33]. At the same time the cytochrome-oxidase activity rose considerably; in the glia, by contrast, the RNA content fell.

Microelectrophoretic analyses showed that the neuronal RNA being formed had changed in composition, containing more guanine and less cytosine [15]; the ratio of adenine to cytosine increased from 0·66 to 0·77. In RNA derived from the isolated nuclei of such nerve cells, the guanine had increased relatively more than in the cytoplasm (table v). The change was towards the 'nucleolar' type of RNA, characterized by high guanine and cytosine content.

A change in the RNA composition was also observed in the neuronal glia, but opposite to that found in the nerve cell. The guanine content fell and the cytosine increased, and the ratio of adenine to cytosine fell from 0·65 to 0·50.

The nerve cells also showed a big increase in RNA one hour after administration of tranylcypromine; this is a monoamino-oxidase inhibitor, and was given in doses of 0·3 mg/kg. The change in composition of the RNA produced consisted in a decrease of the guanine and an increase in the cytosine, the opposite to what was found in the experiments with triap. The same type of changes were found in the glia, although there they were more pronounced. The changes in the RNA composition could first be observed in the glia and subsequently in the nerve cell; this time-sequence may be significant.

These results leave no doubt that synthetic activities are interlinked between the neuron and its glia, even down to the molecular level. They seem particularly significant because the production of RNA is ultimately an expression of the genome of the cell.

A POSSIBLE FUNCTION OF THE GLIA IN THE NEURON-GLIA UNIT

Are the glia more than metabolic appendices to the neuron? Can they also assist the functioning of the neuron by linking together electrical phenomena and metabolic activities? The experimental evidence, briefly recapitulated below, suggests that they can.

TABLE V

Composition of RNA in Deiters' nerve cell nuclei from rabbits injected with 20 mg/kg of tricyano-amino-propene and killed after one hour

	Controls	Tricyano-amino-propene
	Molar proportions	Molar proportions
Adenine ..	21·3 ± 0·42	21·5 ± 0·54
Guanine ..	26·6 ± 0·27	30·0 ± 0·45
Cytosine ..	30·8 ± 0·38	30·4 ± 0·49
Uracil	21·3 ± 0·43	18·1 ± 0·44

Certain chemical differences were found between the nerve cell and its glia. The latter contained less RNA and protein but more free lipids. The glia showed higher respiratory enzyme activities under moderate functional demands and utilized mainly pyruvate and succinate, whereas the nerve cell principally oxidized glutamate. As a result of stimulation the neuron produces RNA and proteins. This seems to be as characteristic of the neuron as its capacity to produce bioelectrical potentials.

Metabolically, there is a pronounced symbiosis between the glia and their neuron. The glia possess a strategic position interposed between the capillaries and the nerve cell. With increasing complexity of the central nervous system in the species the number of glial cells increases relative to that of the neurons [35]. The highest ratio is seen in man.

The glia and the nerve cell are energetically linked together in a coupled system, the units of which can swing between two positions. Such a system provides great stability.

The high respiratory-enzyme activities of the glia at moderate functional demands fall with increasing physiological stimulation, as did the RNA content. In contrast, enzyme activities and production of RNA and protein rose in the neuron, which has high energy demands and utilizes the energy of the electron-transporting system with priority. The glia at the same time partly resorted to anaerobic glycolysis.

Analysis of the base composition of the RNA

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indicated the existence of a coupled mechanism for polynucleotide production in glia and nerve cell. With chemical stimulation, the neuron responded with a prompt synthesis of nuclear (and perhaps cytoplasmic) RNA, the composition of which varied within certain limits. The differences found so far are concerned with guanine and cytosine. Since the composition of the RNA produced can vary according to the type of stimulation, it seems that the proteins produced assist the nervous function. The change in base composition of the glial RNA went in the opposite direction to that of the nerve cell. The glial RNA changes could be observed before those in the nerve cell, and this seems significant.

These are metabolic considerations. Are these biochemical processes, involving production of RNA and proteins, linked with the capacity of the neuron to generate bioelectrical potentials in a system serving the nervous function? Although the evidence is far from sufficient, the following may serve as food for thought.

The nerve impulses contain information in modulated frequencies. The frequency in the neuron is a thousand times higher than in the glia. When increased stimulation effects a change of the frequency in the neuron, that of the glia can be instantaneously changed by a phase-locking mechanism. This triggers the release of specific nucleotides in the glia, which are transferred to the neuron. Because their base composition is opposite to that of the neuron with respect to guanine and cytosine, the glial molecules could act as inhibitors of neuronal repressor RNA [34]. By blocking a repressor RNA in the neuron, enzyme induction and specific protein synthesis could occur to meet the functional demands. The RNA produced in the neuron could partly be used for synthesis of the enzymes serving the coupled Na^+ and K^+ transport at the membrane.

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Charge generation in thunderstorms

B. J. MASON

In 1752 Benjamin Franklin showed that thunderclouds are electrically charged. A number of mechanisms for producing the charge have been suggested, but they have generally not been able to explain completely the observed phenomena. However, recent investigations of thunderclouds by radar and by instruments carried in balloons and aircraft have suggested a new mechanism. This article describes these investigations and gives an account of a new theory, strongly supported by laboratory experiments, that has resulted from them.

It is now more than two hundred years since Benjamin Franklin flew his famous kite and demonstrated the electrification of thunderclouds. A year later, in 1753, he successfully collected electricity from an insulated metal rod exposed to an overhead thunderstorm, stored it in a Leyden jar, and then compared it with a known positive charge from a rubbed glass rod. On the basis of this and later experiments he concluded that 'the clouds of a thundergust are most commonly in a negative state of electricity, but sometimes in a positive state—the latter, I believe, is rare'.

This statement of Franklin's remained the only direct and reliable information on the subject for nearly 170 years—in fact, until C. T. R. Wilson published the first of his classical papers in 1916. But in recent times, and especially during the last fifteen years, the thunderstorm has been explored on an increasing scale from aircraft, and by radar and balloon-borne instruments, and we now have a reasonable working picture of its structure, evolution, and behaviour.

In essence, we can regard the thundercloud as an electrostatic generator that produces electrical charges, both positive and negative, and separates them so that positive charges become concentrated in one region of the cloud and negative charges in another. As separation of the positive and negative charges proceeds, the potential difference between the two oppositely charged regions, or between one of them and the earth, grows until electrical breakdown of the air occurs. At least part of the separated charge disappears during the passage of a lightning flash. More charge is then separated and the same sequence of events is repeated.

The structure and mechanism of the lightning discharge has been described recently in this journal by D. J. Malan [1]. The present article is concerned with the origin of lightning, that is with the mechanisms of charge generation and separation that are fundamental to any solution of the thunderstorm problem. There have, of course, been many theories of these phenomena, but none has withstood the tests of time and the accumulation of detailed information concerning the structure and behaviour of thunderstorms. A satisfactory theory must explain quantitatively how electric charge is generated and separated at a sufficient rate to account for its dissipation in lightning flashes. It must account for the observed distribution of positive and negative charges in the thundercloud and the observed changes in the electric field produced by lightning flashes. The theory must also be consistent with the size and duration of a storm, as well as with the scale and intensity of the rain and hail that accompany the lightning.

THE STRUCTURE OF A THUNDERSTORM

Exploration using aircraft and radar reveals that a thunderstorm consists of one or more active centres—'cells'—that contain strong vertical air currents and that are the locale of raindrop and hailstone formation and of lightning activity. Each cell goes through a fairly well-defined life-cycle of growth, maturity, and decay. The whole life of a cell may last about an hour, and the mature stage, during which precipitation is released and the lightning occurs, lasts for about 15–20 minutes. Very large persistent thunderstorms may arise from successive cells developing

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Was born in 1923 and educated at University College, Nottingham. In 1947 he was appointed lecturer in Meteorology at the Imperial College of Science and Technology. There he established a research group working on the basic physics of cloud- and rain-forming processes. From 1957 to 1961 he was a Warren Research Fellow of the Royal Society, and has recently been appointed by the University of London to the first Chair of Cloud Physics.

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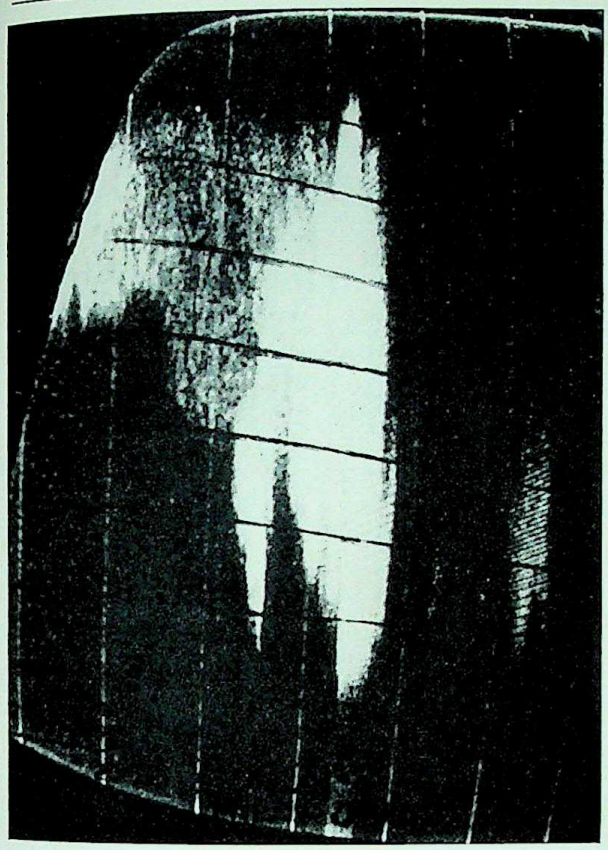

FIGURE 1 – *Radar echoes from a very large thunderstorm. The top of the main echo reaches to about 42 000 ft and is about 15 miles in horizontal extent. A small cell in an early stage of development is seen on the right. Horizontal lines mark height intervals of 5000 ft and the vertical lines mark horizontal intervals of 10 miles. (By courtesy of the Director-General, Meteorological Office, London.)*

in turn; figure 1 shows a radar picture of such a complex storm.

A typical mature cell, or that part of it in which precipitation particles are detected by radar, may be one to five miles across and extend vertically from the cloud base to a level at which the temperature is −40°C, a depth of typically 7·5 km; the visible cloud will usually be surmounted by a vast anvil-shaped mass of ice crystals reaching to even greater heights. The vertical air-currents in a mature thunderstorm may have speeds greater than 30 m/sec, although values of 5–10 m/sec are more common.

The bases of thunderstorms are usually warmer than 0°C, so that, in the early stages of development, water drops will predominate in the lower regions and a mixture of ice crystals and supercooled droplets will appear at colder levels up to the −40°C level, above which height only ice crystals exist. The growth of raindrops by coalescence between water drops of different sizes and speeds of fall starts in the lower part of the cloud, and the capture of supercooled droplets by ice crystals initiates the formation of pellets of soft hail at higher levels. Some of these ice pellets may later develop into true hailstones, but although intense storms giving large hail are almost invariably accompanied by lightning, the presence of large hail does not appear to be a frequent or necessary condition for the onset of lightning. The growth of the precipitation particles continues until their size and concentration become so large that they can no longer be supported by the upward air currents, and they begin to fall towards the earth. The downward rush of heavy precipitation helps to destroy the upcurrent and a downcurrent then starts; this marks the beginning of the mature stage of the storm, with which we shall now be mainly concerned. These features of the thunderstorm are illustrated schematically in figure 2.

ELECTRICAL CHARACTERISTICS OF A THUNDERSTORM

The distribution of electric charge inside a thundercloud was first deduced by C. T. R. Wilson from a study of the manner in which the changes of electric field produced at the ground by lightning flashes varied with the distance from the storm. He concluded that there was a positively charged upper region and a negatively charged region lower down. The general validity of this picture has been amply confirmed by later work, although individual clouds may be more complicated in detail than Wilson's simple bipolar model suggests.

The most direct information comes from measurement of the magnitude and direction of the electric field at different heights in the cloud. The pioneer work was carried out at Kew Observatory just before the second world war by Sir George Simpson and his collaborators [2, 3], with a recording apparatus attached to a balloon. The records showed unmistakably that the upper part of a thundercloud is positively charged and most of the lower part negatively charged. In the majority of storms there was evidence also of a localized positively charged region near the base of the cloud. The upper positive charge P was associated with temperatures that were usually below −20°C and always below −10°C. The temperature at the centre of the negative charge N was usually just below 0°C, and that of the lower positive charge p was above 0°C. In other words, the bulk

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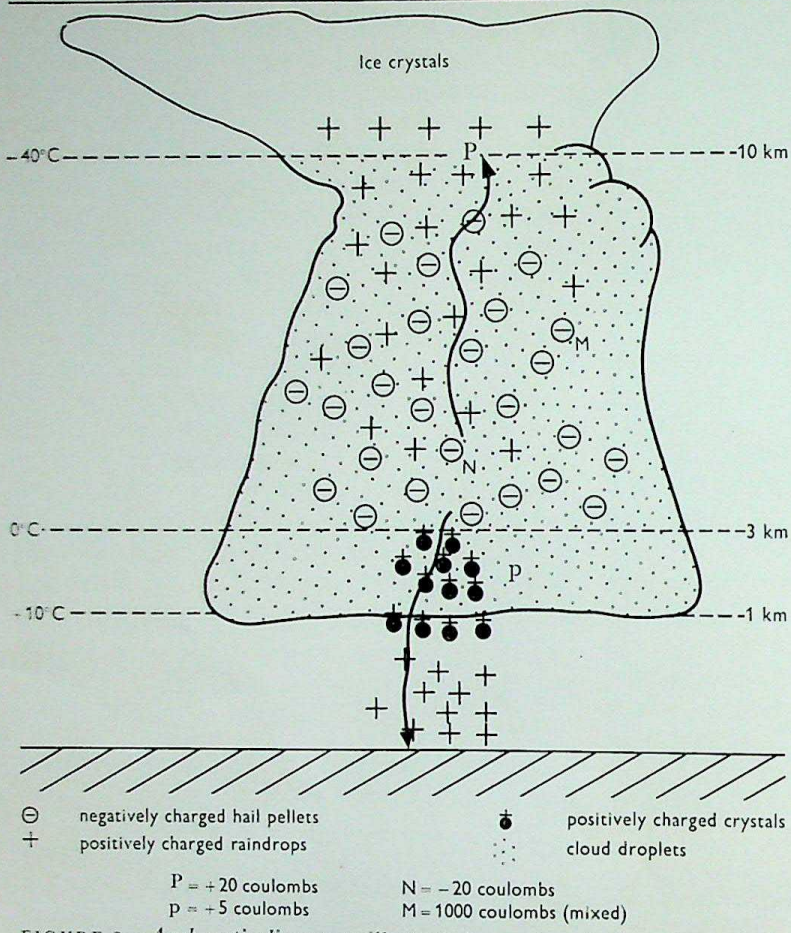

FIGURE 2 – *A schematic diagram to illustrate charge separation in a thunderstorm.*

of the separated charges occurred in regions where the temperature was below 0°C; this fact will weigh heavily when we come to discuss the origin of the charge.

Information on the strength of the electric fields in thunderclouds is rather scanty. Simpson's records rarely showed fields in excess of 100 volts per cm, but this was because sparking occurred at the electrodes of the instrument in the stronger fields that no doubt existed near the electrical centres of the storms. More recent balloon observations in the United States, using telemetering, have occasionally reported fields up to 2000 V/cm. Important information has also been obtained in America by R. Gunn [4], who used aircraft. The mean of the maximum field strengths encountered in nine storms was 1300 V/cm, and on one occasion a field of 3400 V/cm was measured just before the aeroplane was struck by lightning. The most intense fields were encountered near the 0°C level. Such fields, of about 1000 V/cm, are what we should expect to exist near the centre of a single bipolar cloud in which charges of the order of 20 coulombs are separated by distances of some 3 km, and the fields indicate that the total potential difference between the main charge centres may be between 10^8 and 10^9 volts.

Measurement of the change in the vertical component of the electric field at the ground produced by a distant lightning flash makes it possible to calculate the electric moment M destroyed. This is equal to $2QH$, where Q is the charge neutralized and H is the vertical separation of the main positive and negative charge centres for an internal flash, or the height of the negative charge above ground for a discharge to earth. A mean value of M for all types of flash is 110 coulomb kilometres; taking values of H ranging from 2 to 3·5 km for cloud-to-ground discharges and up to 5 km for internal flashes, the charges neutralized are about 10–30 coulombs. As flashes of lightning from an

FIGURE 3 – *The appearance of large spikes in the ice shells of freezing water drops. The shell is ruptured as the liquid interior expands during freezing, and the water is ejected through cracks in the shell to freeze into spikes of this kind. The rupture of the ice shell is also accompanied by the ejection of small positively charged ice splinters.*

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individual thunderstorm cell occur at intervals of about 20 sec, the current dissipated by lightning is of the order of 1 amp.

Negative charge is generally transferred to the earth by cloud-to-ground discharges, but during internal flashes, negative charge invariably moves upwards. Another very important source of charge dissipation is the point-discharge current (St Elmo's Fire) that sets in at pointed objects, such as trees and blades of grass, whenever the electric field exceeds some critical value (usually about 10 V/cm) for the particular object. With a thunderstorm overhead, the point-discharge currents usually transfer positive charge upwards. This tends to neutralize the negative charge in the lower part of the cloud. This current is an important agent in discharging the thunderstorm 'dynamo', which also loses positive charge from its upper pole to the highly conducting upper atmosphere above it, and loses charges of both signs on precipitated droplets and hail. The passage of a lightning flash is accompanied by an almost instantaneous drop in the electrical field strength as measured at the ground. Almost immediately, the field starts to recover, generally rapidly at first and then more slowly. The initial rate of recovery suggests that, if this rate were maintained, the cloud would, on average, become re-charged in about 7 seconds. However, leakage of charge by point-discharge currents, rain, and natural ionic currents increases this time to about 20 seconds, after which a second flash may occur.

The rebuilding of electric moment involves not only a generation of charge but a vertical separation of charges of opposite sign. It is generally believed that the negative charges are attached to the precipitation particles and that the positive charges are carried up on the much smaller cloud particles, so that their rate of vertical separation cannot exceed the velocity (v) of fall of the precipitation particles in still air. Now, the initial rate of regeneration of moment would, if it remained constant, replace that destroyed by the discharge, 110 coulomb kilometres, in 7 sec.

Hence $M = 2QH = 110$ coulomb kilometres; $H_{max} = 7v$; and therefore $Q_{min} = \frac{110 \times 10^3}{2 \times 7v} = \frac{8000}{v}$ coulombs, where v is measured in metres per second. If we take v as 8 m/sec, the magnitude of the charge in the process of being separated immediately after a discharge is about 1000 coulombs. This quantity of charge would be sufficient to supply a number of lightning flashes without replenishment, so that the primary charge-producing mechanism need not necessarily supply new charge to the region as rapidly as it is dissipated by a brief spell of lightning activity. We may imagine that the bulk of the 1000 coulombs is distributed over a considerable volume in the form of mixed positive and negative charges; that separation proceeds throughout the volume to produce a positive region near the top and a negative region near the base, as represented in figure 2; and that a discharge occurs after, say, 20 of the 1000 coulombs have become separated.

REQUIREMENTS OF A THEORY OF THUNDERSTORM ELECTRIFICATION

Having reviewed our present knowledge of the meteorological and electrical characteristics of the thunderstorm, we may say that a satisfactory theory of thunderstorm electrification must be consistent with the following facts:

(*a*) The average duration of precipitation and lightning activity from a single thunderstorm cell is about 30 minutes.

(*b*) The average electric moment destroyed in a lightning flash is about 100 coulomb kilometres, and the corresponding charge is 20–30 coulombs.

(*c*) The magnitude of the charge which is being separated immediately after a flash by virtue of the falling speed of the precipitation elements is about 1000 coulombs.

(*d*) In a typical thunderstorm cell this charge is generated and separated in a volume bounded by the 0°C and −40°C levels with a typical radius of 2 km, and therefore a volume of about 50 km^3.

(*e*) The negative charge is centred near the −5°C level and the main positive charge is situated some kilometres higher up; a subsidiary positive charge often exists near the cloud base where the temperature is usually a little above 0°C.

(*f*) The charge generation and separation processes are closely associated with the development of precipitation.

(*g*) Sufficient charge must be generated and separated to supply the first lightning flash within 10–20 minutes of the first appearance of precipitation particles large enough (greater than about 0·1 mm radius) to be detected by radar.

In round figures, the requirement is to generate about 1000 coulombs of charge in a volume of

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about 50 km³ in a period of about 20 minutes, that is at an average rate of 1 coulomb/km³ per min.

The various mechanisms that have been suggested in the past have been examined by the author in the light of this requirement [5] and appear either incapable of producing charge at anything like this rate or incompatible with modern knowledge of the structure and behaviour of thunderstorms.

SEARCH FOR A NEW THEORY

In searching for a new mechanism, the author was particularly influenced by the following facts. The main charge centres appear at levels in the cloud where the temperature is below 0°C; it is therefore natural to associate charge generation with water in either the supercooled or solid state. Indeed, J. Kuettner [6], from observations made inside thunderclouds capping the Zugspitze in Germany, reported that the onset of lightning coincided with the appearance of solid precipitation in the form of pellets of soft hail. And, very recently, cloud physicists at the University of Chicago have recorded sudden large increases in the electrical field when they observe soft hail from their aircraft.

Laboratory experiments performed by W. Findeisen [7] in Germany during the last war, and repeated by other workers since, have shown that when supercooled droplets are sprayed on a cold surface to produce a layer of rime ice, the rime acquires a charge. The air also acquires a charge, but of opposite sign. Most of the workers reported that the charge acquired by the rime was negative.

In some recent experiments, B. J. Mason and J. Maybank [8] have found that the freezing of water droplets with a radius of 0·03–1·0 mm is often accompanied by the bursting of the ice shell (see figure 3) and the ejection of small splinters of ice carrying a positive charge, leaving negative charges on the residues of the droplets. This suggests that the formation of pellets of soft hail in a cloud by the impaction and freezing of supercooled drops will lead to the formation of negatively charged hailstones that will fall towards the base of the cloud while the positively charged ice-splinters are carried by the upcurrents towards the top of the cloud, as shown in figure 4.

A rather different mechanism has been suggested by S. E. Reynolds, M. Brook, and M. F. Gourley [9] in the United States. They have obtained laboratory evidence that a hail pellet can be given a charge by the impact and rebound of small ice-crystals having a slightly different temperature. They found that a slightly colder crystal bounced off with a positive charge, leaving the hailstone negatively charged; a reversal of the temperature-differential produced a reversal of signs. Under natural conditions, one would expect the surface of the hailstone to be slightly warmer than the ice crystals because of the latent heat released by the droplets freezing on the hailstone; the hailstone would thus be expected to acquire a negative charge.

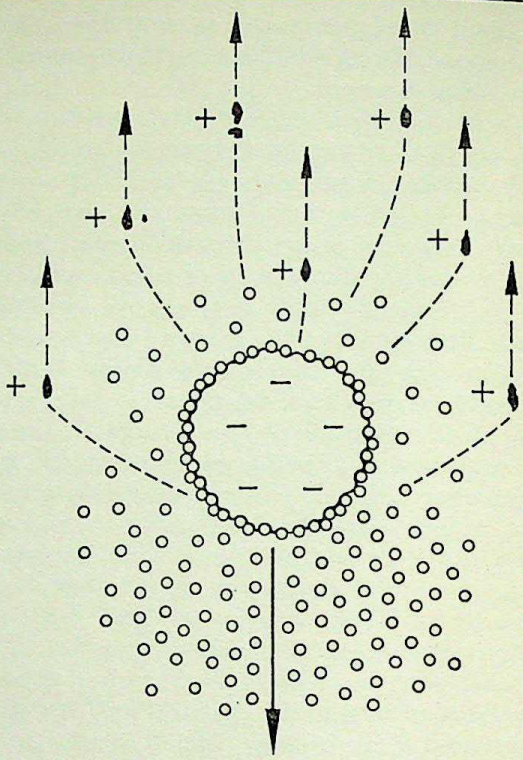

FIGURE 4 – *Diagram illustrating the growth and negative charging of a hail pellet as it falls through a cloud of supercooled droplets. Droplets that impinge on the hail pellet freeze and eject small positively charged ice splinters, which are carried towards the top of the cloud in the upcurrents. The heavier hailstones carry a negative charge towards the cloud base.*

The hailstone electrification caused by both the splintering of freezing droplets and collisions with ice-crystals has recently been the subject of a detailed study by J. Latham and B. J. Mason [10, 11]. The results of these experiments will now be described, together with a new theory of the mechanism involved.

BASIC MECHANISM OF CHARGE SEPARATION

The fundamental problem is to explain how the electric charge can be separated during the bursting of a freezing drop or during a temporary

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contact between two pieces of ice at different temperatures. In both cases the existence of a temperature gradient is involved, and the new theory discusses how the hydrogen and hydroxyl ions formed by the dissociation of a small fraction of the ice molecules become separated under the influence of this temperature gradient.

The process depends essentially on two facts. One is that the concentrations of positive and negative ions increase quite rapidly with increasing temperature; the other, that the hydrogen ion diffuses much more rapidly through the ice crystal than does the hydroxyl ion [14]. Thus, if we imagine a steady temperature difference maintained across a piece of ice, the warmer end will initially possess higher concentrations of both positive and negative ions. The more rapid diffusion of H^+ ions down this concentration gradient will lead to a separation of charge, with a net excess of positive charge in the colder part of the ice. The theory indicates that a steady temperature difference of ΔT (°C) will be accompanied by a potential difference of about $2\Delta T$ (millivolts) across the ice. Such potential differences have been measured experimentally across specimens of pure ice, and it has been shown that they are not markedly affected by the presence in the ice of such impurities as dissolved salts and gases.

It follows that, when two pieces of ice at different temperatures are brought into temporary contact and separated, the warmer piece should acquire a negative charge and the colder one should acquire an equal positive charge. The theory indicates that there should be a maximum charge difference of $3 \times 10^{-3}\Delta T$ e.s.u. between each square centimetre of contacting surface when the surfaces are separated after about 0·01 sec. If they are left in contact for longer times, the charge separation will be decreased as the two pieces of ice become more nearly equal in temperature.

These conclusions have been confirmed by experimental measurements that show that very little charge separation occurs if the contact period exceeds 0·5 sec. Experiments in which the ice was contaminated with carbon dioxide, hydrofluoric acid, and sodium chloride, in concentrations up to fifty times that normally present in rainwater, showed that the electrification was not greatly influenced by these impurities.

The preferential migration of protons down a temperature gradient explains why an ice crystal making temporary contact with a warmer hailstone will leave the latter with a negative charge. The electrification of freezing water droplets can also be explained by this mechanism, in the following manner. During the early stage of freezing, a shell of ice is formed round the outside of the drop and the liquid interior proceeds to freeze at a rate determined by the rate of dissipation of the latent heat of fusion to the surroundings. During this process a radial temperature gradient is established across the ice shell, the inner surface being held at 0°C by the water that is still liquid inside, the outer surfaces cooling towards the air temperature. According to the above theory, protons will migrate preferentially down this temperature gradient and produce a positive charge in the outer layers of the ice. When the droplet bursts by the expansion of the centre as it freezes, splinters ejected from the outer layer will tend to carry away a positive charge and leave the remainder negatively charged. This corresponds to the experimental observations.

CHARGING OF HAILSTONES

In order to investigate the relative importance of these two processes—the freezing and fragmentation of supercooled droplets, and the bouncing of ice crystals on the surface of hailstones—Latham and Mason [11] have measured in the laboratory the rate at which artificial hailstones become charged by the impaction of small ice crystals and supercooled water droplets. The experiments were conducted in a cold-room at temperatures down to −16°C. The simulated hailstone (a small sphere of ice) was held stationary in a wind tunnel as ice crystals 0·05–0·02 mm diameter or supercooled droplets 0·02–0·1 mm diameter were blown past it at speeds varying from 1 to 30 m per sec. This range covers the speeds at which most hailstones fall through clouds. It was possible to vary the temperature of the hailstone relative to that of the surrounding air. Temperatures were measured with thermocouples and the charge acquired by the hailstones was measured with a sensitive electrometer.

It was found, as might have been expected, that the sign of the electrical charge produced by collisions between ice crystals and hailstones was governed by the temperature difference between them; the hailstone became negatively charged when it was warmer than the rebounding crystals, and *vice versa*. The magnitude of the charge was proportional to the temperature difference but rather insensitive to the size and impact velocity of the crystals. With a temperature difference of 5°C, a rebounding crystal having a diameter of about 0·05 mm produced charges averaging 5×10^{-9}

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e.s.u. At temperatures within a degree or two of the melting point, the crystals stuck to the surface of the hailstone and the charging ceased.

When supercooled droplets were used in this experiment, the growth of the hail pellet by the accretion and freezing of supercooled droplets was accompanied by the ejection of small splinters of ice and the hailstone acquired a negative charge. Droplets less than 0·03 mm in diameter produced a few splinters and little charge; when the size of the droplets was increased, the rates at which both splinters and charge were produced increased until large drops moving at higher speeds caused splashing on the surface of the hailstone. The charging was then greatly reduced and even reversed. In a typical experiment at an air temperature of $-15°C$, droplets of 0·08 mm diameter striking at 10 m per sec froze to produce, on average, twelve ice splinters and a charge of 4×10^{-6} e.s.u. per drop. As before, these results were not appreciably affected by the presence of contaminants in the water in concentrations that are usually found in rainwater.

APPLICATION OF LABORATORY RESULTS TO THE THUNDERSTORM

The results of these laboratory experiments can be used to calculate the probable rates of charge generation that will be associated with the formation of soft hail in a typical thunderstorm. Aircraft measurements provide fair estimates of the number of hail pellets, ice crystals, and supercooled cloud droplets to be found in a given volume of cloud. One can therefore calculate the rate at which the more rapidly falling hailstones will collide with both ice crystals and supercooled droplets. Latham and Mason's laboratory measurements of the charge produced by each such collision can then be used to compute the rate at which the hailstones will acquire negative charge by both processes. Finally, one can calculate the rate at which the charge and the electric field caused by separation of the charge build up. The results are as follows. While the ice-crystal–hailstone collisions appear incapable of generating charge at the required rate, the impaction, freezing, and splintering of the larger supercooled droplets on the surfaces of hail pellets can readily generate charge at the required rate (1 coulomb/km^3 min) and create electric fields of several thousand volts per cm in about 10 minutes [15].

The author therefore believes this to be the principal mechanism of thunderstorm electrification, and believes that it depends essentially on a fundamental thermo-electric property of ice, the preferential migration of protons in ice under the influence of temperature gradients.

ELECTRIFICATION OF CLOUDS WARMER THAN 0°C

Although the main charge-centres of large thunderstorms appear always to be located in the sub-freezing part of the cloud and can be explained in terms of the hailstone-riming mechanism just described, there are on record three or four reports of lightning being observed from clouds whose tops were warmer than 0°C and could therefore have contained no ice. Certainly lightning from such warm clouds appears to be a rare phenomenon, although rain falls frequently from such clouds in warm maritime areas like Hawaii and the Caribbean. The few reports that we have of lightning from warm clouds were made from aircraft flying over the Caribbean ocean in twilight or darkness and are not entirely convincing. This is because a cloud spotted 100 miles away may well subside before an aircraft reaches it, and lightning flashes may sometimes travel over horizontal distances of 50–100 miles in extensive thundercloud systems. But, should the reports be substantiated by detailed observations that firmly establish that lightning may actually originate inside isolated clouds whose tops do not, at any stage, extend well above the 0°C level, it will be necessary to look for an additional process of electrification.

It would be convenient to find a mechanism that would also explain the appearance of the lower subsidiary positive charge that appears in the bases of most thunderstorms at temperatures above 0°C. This charge, which is associated with the region of heaviest rain, is usually attributed to a stream of positive ions. This is produced by corona (point) discharge at the earth's surface by the thunderstorm's own electric field and then streams upwards towards the base of the cloud in an attempt to neutralize the main negative charge. The raindrops are believed to acquire a positive charge by capturing some of these ions. Beneath the most intense parts of a storm this point-discharge current may reach typical values of 0·02 amp/km^2, so that if it persists for 10 minutes a total positive charge of 10–20 coulombs may flow into the base of the cloud. Interception of a part of this by the raindrops might then account for the lower positive charge, which Simpson estimated to be about 4 coulombs. But this point-discharge current, being initiated and maintained by the electric field of the main charge-centres, must be regarded

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as a consequence rather than a cause of the cloud electrification, and it is therefore difficult to see how it could account for the main electrification of warm clouds over the oceans where, in any case, one would imagine the point-discharge currents to be much smaller than over land.

What appears at first sight to be a more attractive mechanism for charging a cloud composed wholly of liquid-water drops is the one suggested as early as 1913 by J. Elster and H. Geitel [12] and recently revived by J. D. Sartor [13]. This considers the electrical charging of raindrops as they fall in a vertical electric field and collide with the much smaller cloud-droplets that they overtake. The raindrop will be electrically polarized by the field as shown in figure 5; in a downwardly directed field such as exists in the atmosphere in fine weather, the lower half of the drop will carry a positive charge and the upper half an equal negative charge. Elster and Geitel suggested that, after collision, some of the cloud particles would rebound from the underside of the raindrop, carry away some of its positive charge, and leave the raindrop with a net negative charge. The falling drops, in carrying their negative charge towards the base of the cloud, would enhance the original field, and so the whole electrification process would build up rapidly. A detailed mathematical treatment of the problem shows that if only one per cent of the cloud droplets striking the lower half of the raindrops were to bounce off, and if this percentage were independent of the electric field strength, the field would grow exponentially with time and, in a cloud producing rain at the rate of 1 cm/h, would reach 1000 times its initial fine-weather value in only 10 minutes.

Now laboratory experiments indicate that, in the absence of an electric field, the fraction of cloud droplets that actually rebound after striking raindrops is small; the great majority of collisions result in coalescence. However, no experiment sufficiently accurate to detect non-coalescence to

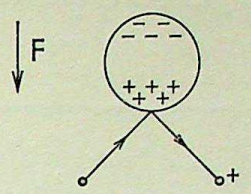

FIGURE 5 – *Diagram to illustrate how a raindrop, polarized in an electric field, may become negatively charged if cloud droplets rebound from its lower surface.*

the extent of only a few per cent has yet been performed. This problem, which becomes complicated further by the presence of electric fields and of free charges on the drops, is now being studied by the author. It has been found that although droplets of 0·1 mm diameter may rebound after striking an uncontaminated plane surface of water or a much larger drop, they can always be made to coalesce by applying vertical fields of only about 10 volts per cm. The droplets become distorted as they approach the water surface, small protuberances develop at their ends in the direction of the field, and complete coalescence occurs, probably because small electric discharges occur at the protuberances and cause rupture of the intervening air film. This suggests, perhaps, that once fields of about 10 V/cm are established in the cloud, coalescence between colliding cloud droplets and raindrops is assured. In this case the Elster-Geitel charging mechanism will cease long before fields strong enough to initiate lightning are produced.

At the present time we are unable to suggest a mechanism that would convincingly account for the origin of lightning in clouds consisting wholly of liquid water. But there is strong evidence in favour of the riming-hailstone mechanism being the dominant one in the typical large thunderstorm. However, it would be unwise to assume that there is only one mechanism of charge generation. The thunderstorm, which has shown great reluctance to yield its secrets, may well have further surprises.

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The biochemistry of fruit maturation

J. B. BIALE and R. E. YOUNG

The physiology and biochemistry of the ripening of fruit are interesting in their own right; in addition, many of the processes involved occur in the ageing of all living organisms. This article describes studies of the rates of oxidation in plant cells, of the hormone-like substance that initiates fruit formation, and of the effects of light and of gases on the speed of maturation of cells. Modern methods of fruit storage are based on biochemical and physiological studies of ripening; this makes the study of commercial, as well as academic, interest.

Research on fruits has been prompted by their commercial importance and nutritive value, and by the realization that biological processes that are common to all living organisms can be studied in fruits. The structural and functional transformations that occur in cells during ripening are particularly interesting to the fruit physiologist.

There are several good reasons for using fruits in a study of the general biological problem of ageing. The fruit is capable of prolonged independent existence even after removal from the parent plant; as long as it is kept in a humid atmosphere, it requires no external supply of water to maintain the cells in a turgid condition. Most fruits, at least the ones eaten by man, are amply supplied with respiratory substrates; in the ordinary sense, starvation is of little or no consequence, although the depletion of certain specific substances required in small amounts and not synthesized in detached fruits may play a role in the metabolic pattern. The study of fruits avoids complexities resulting from the interplay of several processes, such as happens with photosynthesis and respiration in leaves.

The central reactions in ripening are biological oxidations. This term is used advisedly to include glycolytic as well as respiratory activities. The former refers to the conversion of carbohydrates to pyruvic acid in the presence or absence of oxygen. The pyruvic acid is converted in the absence of oxygen to fermentative end-products such as alcohol, lactic acid, and propionic acid. The term respiration is reserved then for the reactions requiring oxygen for the enzymatic oxidation of carbon compounds to carbon dioxide and water.

The fruit physiologist has applied the techniques that have been so successfully developed for the study of intermediary metabolism to that of the ripening of fruit. In turn, he has contributed to the elucidation of the biosynthetic pathways of substances formed in fruits, such as ascorbic acid, pectins, anthocyanins, and ethylene. The present state of knowledge in biochemical fruit physiology will be discussed here in terms of results obtained from the whole fruit, the tissue disc isolated from the fruit, and the subcellular components. Most research has been with fully mature fruits [1, 2, 3]; however, the physiology of developing fruits is important [5], and will also be briefly described.

PHYSIOLOGICAL ASPECTS OF FRUIT DEVELOPMENT

Physiologically, it is immaterial whether the fruit is formed from an enlarged ovary wall, from the receptacle (the tissue supporting the ovaries), or whether it is the product of the fusion of floral parts with ovarian tissue. Typical examples of each process are shown in figure 1. More interesting than the morphogenetic origin is the fact that the development of the fruit needs a hormonal stimulus. This may be supplied by pollen, pollen

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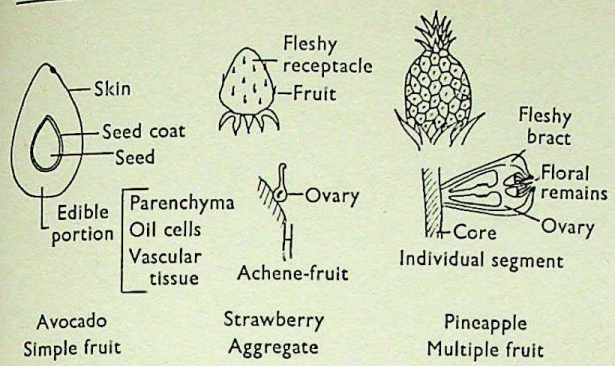

FIGURE 1 – *Diagram of three different morphological types of fruits.*

extracts, ovules, or developing seeds (figure 2). The chief source of the hormone appears to be the endosperm, the tissue within the seed that results from the fusion of a pollen nucleus with two nuclei of the ovule. Attempts to identify the hormones have met with some difficulty owing to the presence of growth inhibitors in the extracts. These inhibitors interfere with the bio-assay, which depends on the measurement of plant growth caused by substances produced in quantities too minute to be determined by physico-chemical methods. However, there seems little doubt that hormones are involved.

Once the ovary has been stimulated, it grows by one of the following patterns: (*a*) single sigmoid growth curve, as in the apple and tomato, or (*b*) double curves, exemplified by the cherry, fig, and some berries. In the second group, rapid growth takes place shortly after fertilization and during a brief period between maturation, with a plateau between the two cycles. The discontinuity in the growth curve has been traced to competition for auxins between the developing fruit and the growing embryo. In general, however, growth by cell division is of relatively short duration, while cell expansion may continue until maturity. The size of the fruit is determined by the number of cells formed as well as by cell size.

During the early stages of development, the cells consist mostly of protoplasm. With cell enlargement, vacuoles are formed; carbohydrates and other compounds that have been formed in the leaves accumulate in the cells and dilute the proteinaceous materials; and substances specific to the particular fruits are formed. The composition of mature fruits is very varied. Some fruits, such as the apple, banana, and date, accumulate carbohydrates; the avocado and the olive are typical fat-storing fruits. The predominant acid in some fruits is malic (apple, pear), in others citric (citrus, pineapple), and in still others tartaric (grape). In all fruits the protein content is rather low, but it varies from 1·7 per cent (of fresh weight) for the avocado, to 0·3 for the apple. The content of phosphorus, a key element in metabolism, does not vary as widely as does that of protein nitrogen. Phosphorus is present in various organic compounds such as nucleotides, coenzymes, and phosphorylated sugars and acids. The role of some of these in fruit metabolism will be discussed below. Despite the pronounced variations in composition, the patterns of physiological behaviour of fruits of widely differing origin and of different climatic adaptability are few and rather similar.

THE RESPIRATORY PATTERNS OF THE INTACT FRUIT

Fruits, in common with all aerobic organisms, absorb oxygen and give off carbon dioxide. Normally the measurement of the change in partial pressure of either of these gases gives an index of respiratory activity. If the rate of oxygen uptake or of carbon dioxide evolution is followed from the early stages of fruit development until maturation, a continuous decline is observed. The drop is most striking when the results are calculated on the basis of fresh weight or dry weight, but it is more gradual when calculated on a nitrogen basis, because the relative protein content decreases with the formation of vacuoles and the accumulation of sugars, acids, and polysaccharides.

When a fully developed but unripe specimen of a

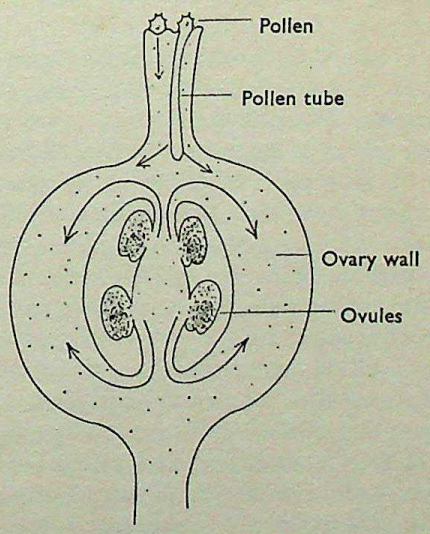

FIGURE 2 – *Sources of hormonal stimulus for fruit development.*

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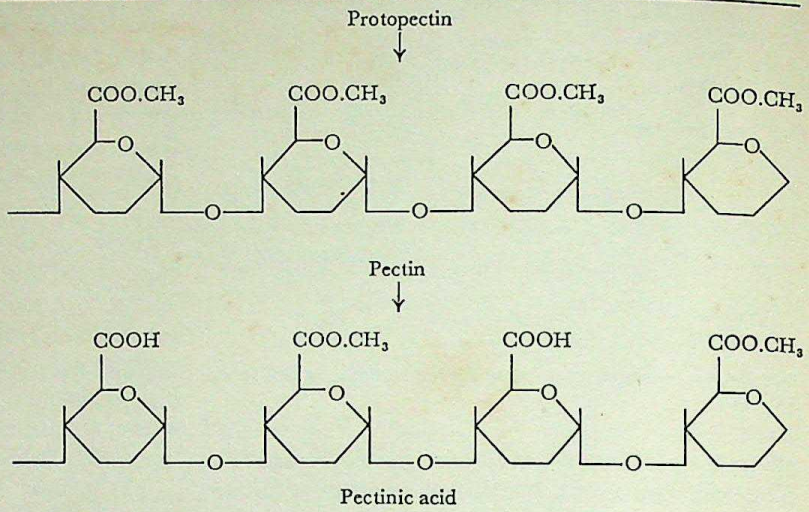
fruit such as the avocado or banana is harvested and placed under conditions favourable for ripening, the rate of respiration continues to decline until a minimum is reached after several days or weeks, depending on state of maturity, temperature, and gaseous environment. Thereupon a sudden and sharp upsurge in respiration ensues. This has been called, by the English investigator F. Kidd, the 'climacteric rise' in respiration. The discovery of the climacteric rise had a pronounced effect on research in fruit physiology. The climacteric may be looked upon as that stage in the life of fruits that separates development and maturation from breakdown. Marked changes in resistance to disease, in cellular properties, and in metabolic reactions occur during this stage and lead to breakdown. Certain readily discernible changes are associated with the climacteric. The disappearance of chlorophyll from the peel of ripening bananas (figure 3) is closely linked to the course of the respiratory climacteric. Similar observations were made on the transition from green to yellow in certain varieties of apple, pear, and papaya, and that from green to red in plums and mangos. The colour changes in most of these cases are due to chlorophyll destruction; the yellow plastid pigments, carotene and xanthophyll, remain relatively constant. Softening of flesh is also closely linked with the respiratory pattern. The avocado, for example, is never ripe enough to eat before the climacteric peak. Some of the chemical changes that occur during the course of ripening are shown in table I.



If one group of substances had to be designated as the most typical of those in ripening fruits, pectins would certainly deserve first consideration. Pectins are made up of chains of galacturonic acid units; galacturonic acid is frequently associated with the sugars galactose and arabinose. Pectin changes during ripening may be depicted as above.

The COOH group of the galacturonic acid residues may be free or esterified with methoxyl groups. The nature of the pectin depends on the ratio of free to esterified carboxyl groups in the chain as well as on the chain length of polygalacturonic acid polymer. Calcium also reacts with carboxyl groups to yield a highly insoluble pectin known as calcium pectate, which is present in the walls between cells. It has been suggested that pectins become soluble in some fruits during ripening by the removal of calcium and not by shortening of the polygalacturonide chain. However, it has been shown in several fruits that both pectin methyl esterase, the enzyme that splits off the methoxyl groups, and polygalacturonase, responsible for splitting the chain, are both active during ripening. If these enzymes are the key to the onset of the ripening process, it remains to be shown how their formation is related to the respiratory upsurge. Alternatively, the enzymes may be present at all times but rendered inactive in unripe fruit by inhibitors. If this is the case, the fate of the inhibitors is worthy of study.

The respiration values at the climacteric minimum and peak are characteristic features for a particular species under a given set of conditions. Not all fruits exhibit a respiratory upsurge. In some, such as lemons, there is a gradual decline in oxygen uptake or carbon dioxide evolution throughout maturation and into senescence. Apparently, no marked protoplasmic or chemical transformations take place to alter the course of the steady decline. The slow utilization of soluble sugars rather than hydrolysis of insoluble carbohydrates characterizes the nonclimacteric fruits. In table II comparative values are given for the rates of respiration of fruits in each of the two classes. The differences between the various kinds are probably due to a combination of factors such as protein content, and the availability of catalytic quantities of organic acids, nucleotides, and coenzymes.

THE REGULATION OF RIPENING

Respiration and ripening of harvested fruits are

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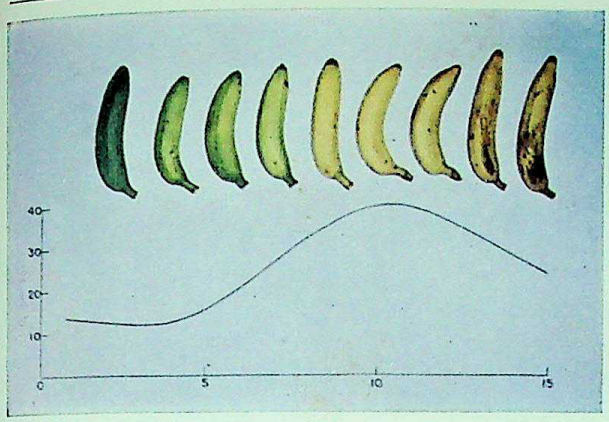

FIGURE 3 – *Climacteric rise of respiration in relation to colour changes in the banana.*

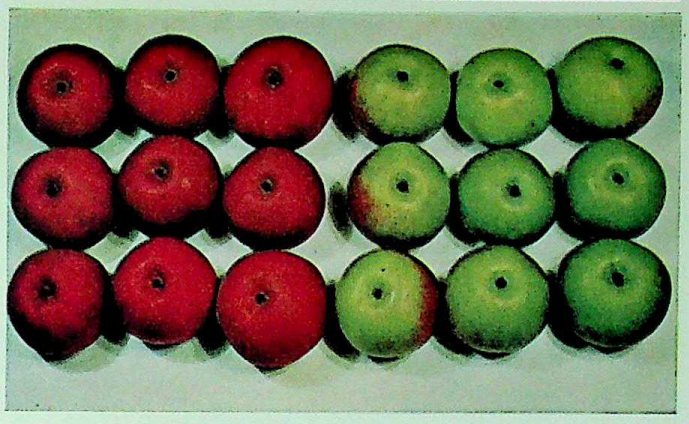

FIGURE 4 – *Effect of light on anthocyanin formation in apples. Left (red) exposed to fluorescent light continuously; right, held in darkness. (Photograph by courtesy of Dr H. W. Siegelman.)*

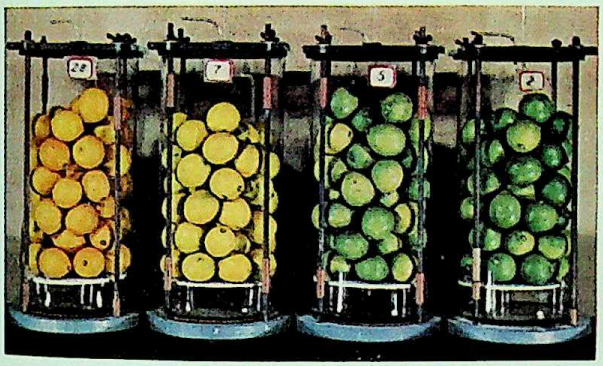

FIGURE 5 – *The effect of oxygen tension on colour changes in lemons. Oxygen concentrations (from the left):* 21% *(air),* 10%, 5%, 2·5%.

FIGURE 6 – *Colour changes in lemons as affected by the interaction of carbon dioxide and ethylene (from the left):* 21% O_2 *(air);* 21% O_2, 10 *p.p.m. ethylene;* 21% O_2, 5% CO_2; 21% O_2, 5% CO_2, 10 *p.p.m. ethylene;* 21% O_2, 10% CO_2; 21% O_2, 10% CO_2, 10 *p.p.m. ethylene.*

controlled by temperature and by gases. Temperature effects are both quantitative and qualitative. Within a relatively narrow range of about 5–30°C, the intensity of respiration in fruits, as in all living organisms, responds to temperature in a manner characteristic of chemical reactions. The increase in rate with rise in temperature is relatively high, though not uniformly so throughout the entire range. From figure 9 it can be seen that the respiration of avocados is more sensitive to temperature than that of citrus. The results and the evidence from experience indicate that, to achieve the best quality, fruits such as the avocado, banana, and pear should be ripened after harvesting at a temperature close to 20°C. The respiratory patterns of avocados kept at several temperatures are illustrated in figure 10. It is clear that at temperatures of from 10°C to 25°C, there was a definite climacteric, and the fruit ripened normally. The only difference between these samples was the time required for ripening. However, a striking difference is noted in the respiratory pattern of avocados held at 5°C and at 30°C. In both cases, no climacteric occurs; the fruit fails to ripen; the tissue darkens; and the fruit is inedible. The effect observed in avocados at 5°C is known as chilling injury and is common to most fruits. Each fruit has its characteristic temperature below which damage will occur. Bananas, for example, will not tolerate temperatures below 12–13°C. Some varieties of apples will store well as low as 1°C, but other varieties should not be kept below 4–5°C. High-temperature injury has been studied for only a few fruits, but there are undoubtedly characteristic temperature-limits for each fruit. It is noteworthy

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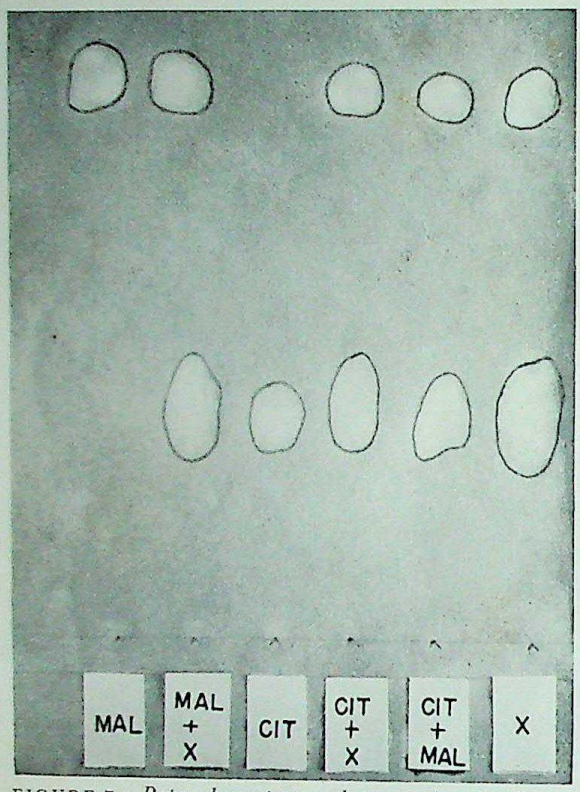

FIGURE 7 – *Paper chromatogram demonstrating the identity of malonic acid from lemon rind. Mal, malonic acid marker. Cit, citric acid marker. X, spot of one fraction from the ion-exchange column shown by this chromatogram to contain both malonic and citric acids.*

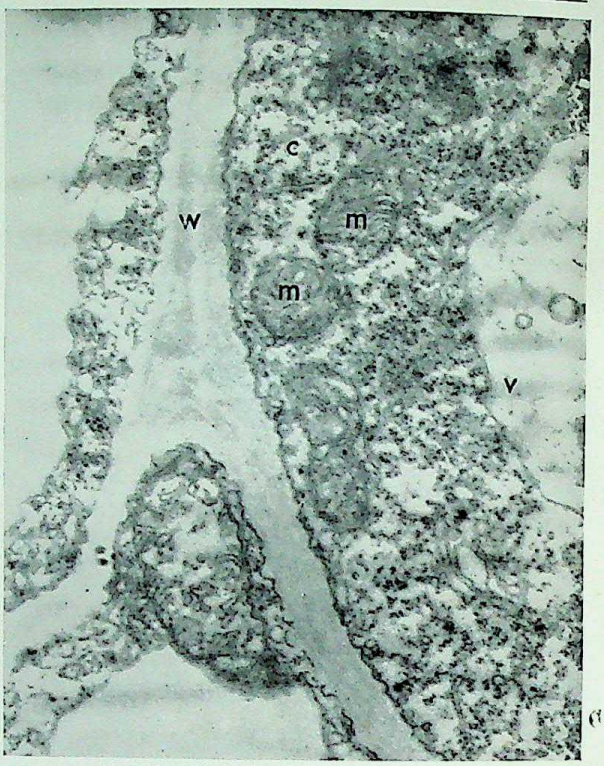

FIGURE 8 – *Mitochondria (m) in hypodermal cells of young unripe apple fruit. The mitochondria have the typical mitochondrion structure consisting of cristae that arise from the double mitochondrial membrane. Other details shown are the cell wall (w), the cytoplasm (c), and vacuole (v). Magnification* ×26 000; *OsO_4 fixation. (By courtesy of Joan M. Bain and F. V. Mercer.)*

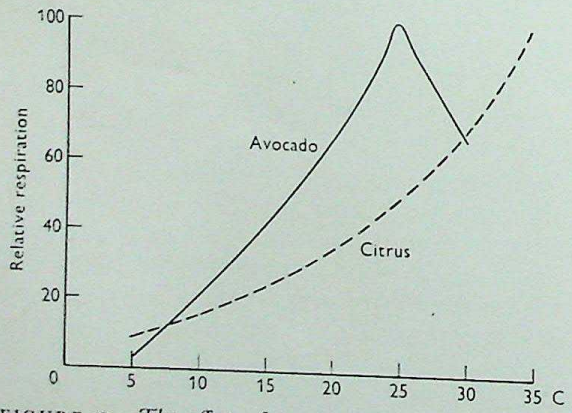

FIGURE 9 – *The effect of temperature on respiration of avocado and citrus fruits.*

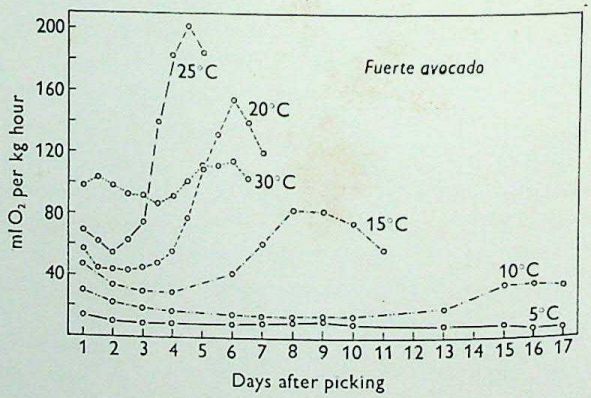

FIGURE 10 – *The effect of temperature on the climacteric pattern of respiration of avocados.*

that many tropical and subtropical fruits are subjected to rather high temperatures while on the tree, but become sensitive to high temperature only when detached from the tree. Injuries caused by low or high temperature are poorly understood from the biochemical point of view.

For many years fruits have been stored in air, but physiological investigations have suggested the use of other atmospheres. Since oxygen and carbon dioxide are involved in the process of respiration, one might expect the partial pressures of these gases and their rates of diffusion to exert a pronounced effect on fruit metabolism. In figure 11, the carbon dioxide production is plotted as a

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TABLE I

Salient chemical changes during ripening in selected fruits[1]

Fruit	Constituent	Fruit part	Grams per 100 grams fresh weight	
			At picking	After storage
Banana	Reducing sugars	Pulp	0·24	15·34
	Non-reducing sugars	Pulp	0·62	2·60
	Starch	Pulp	20·65	1·21
	Acid (as malic)	Pulp	0·40	0·50
	Ascorbic acid	Pulp	0·0053	0·0111
	Protopectin	Pulp	0·53	0·22
	Soluble pectin	Pulp	0·27	0·40
	Chlorophyll	Peel	$1·03 \times 10^{-2}$	None
	Carotene	Peel	$2·0 \times 10^{-4}$	$3·6 \times 10^{-4}$
Apple	Reducing sugars	Pulp	4·7	7·0
	Non-reducing sugars	Pulp	2·8	0·4
	Starch	Pulp	2·0	0·1
	Acid (as malic)	Pulp	1·00	0·60
	Ascorbic acid	Pulp	0·21	0·07
	Protein	Pulp	0·20	0·24
	Protopectin	Pulp	0·68	0·08
	Soluble pectin	Pulp	0·17	0·45
	Chlorophyll	Peel	$2·2 \times 10^{-4}$	$1·5 \times 10^{-4}$
	Carotene	Peel	$3·3 \times 10^{-5}$	$8·0 \times 10^{-5}$
Pear	Protopectin	Pulp	0·80	0·10
	Soluble pectin	Pulp	0·20	0·60
	Acid (as malic)	Pulp	0·32	0·32
	Ascorbic acid	Pulp	0·10	0·06
Avocado	Total sugars	Pulp	0·40	0·05
	Fat	Pulp	21·3	22·3
	Protein	Pulp	1·8	2·0
Lemon	Total sugars	Pulp	9·6	7·7
	Acid (as citric)	Pulp	5·58	5·38
	Ascorbic acid	Pulp	0·067	0·045

[1] The values vary with fruit variety as well as storage conditions and are presented only to indicate the magnitude of changes associated with ripening in selected cases.

function of the oxygen concentration in which the fruit were stored. In the avocado, low levels of oxygen (below 2·5 per cent) suppressed the climacteric pattern and prevented normal ripening. In a gas mixture containing 2·5 per cent oxygen, the onset of the climacteric was delayed, the slope was more gradual, and the time from picking to softening was twice as long. Air enriched with oxygen gave only slightly higher respiration, indicating relatively good conditions for gas diffusion within the fruit. In contrast, the respiration of oranges and lemons increased as a function of changes of oxygen concentration between 5 and 100 per cent. In this case a critical level of 2·5–5·0 per cent oxygen, at which carbon dioxide evolution was at a minimum, was noted. The increase in the respiratory quotient (ratio of carbon dioxide

TABLE II

Comparative rates of respiration of representative fruits at 20°C

Climacteric type fruit			Non-climacteric type fruit	
Fruit	ml O_2 or CO_2/kg hour		Fruit	ml O_2 or CO_2/kg hour
	Min.	Max.		
Apple ..	8	15	Grape ..	12
Avocado ..	35	155	Lemon ..	10
Banana ..	20	60	Orange ..	13
Pear ..	12	33	Strawberry	65

Min. refers to value immediately preceding climacteric rise.
Max. refers to rate at climacteric peak.
For non-climacteric fruit, figures are for steady-state rates.

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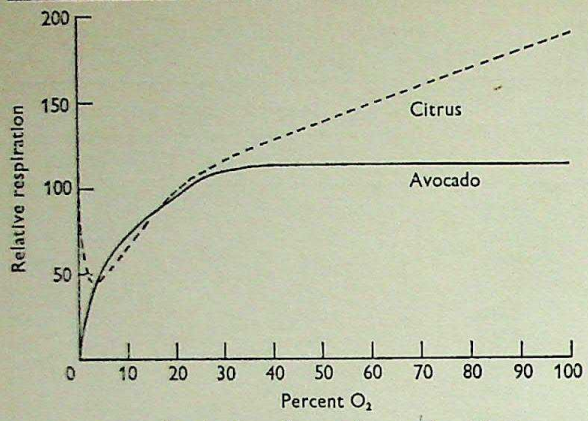
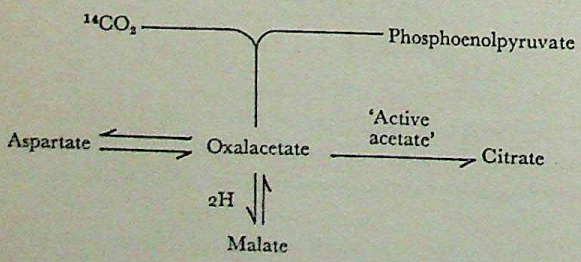


FIGURE 11 – *Respiration of avocado and citrus fruits as a function of the oxygen concentration of the storage atmosphere.*

evolved to oxygen absorbed) indicated that fermentative reactions took place at oxygen concentrations lower than the critical concentration. The oxygen level of 5 per cent was suitable for prolonging the storage life of both avocados and citrus by a factor of two or more. Colour changes of lemons in relation to oxygen concentration are shown in figure 5.

Further benefits in storing fruits have been gained by adding carbon dioxide to air with a relatively low oxygen content. In general, partial pressures equivalent to 5 and 10 per cent carbon dioxide tend to suppress the intensity of respiration. An exception to this trend was found in the case of lemons. Here the exposure to 10 per cent carbon dioxide in combination with 10 or 21 per cent oxygen brought about a stimulation of the oxygen absorption. In order to investigate this unexpected response, lemons were placed for a short time in an atmosphere containing radioactive carbon dioxide. After an exposure of 5 minutes the carbon dioxide was rapidly swept out of the container, and the rind separated and ground in 70 per cent alcohol in a blender. The extract was freed of lipids and analysed chromatographically. It was found that malic, citric, and aspartic acids were labelled rapidly. This suggests that oxalacetate, which is rather labile and evades identification, is the first product to receive radioactivity, following this scheme:



The acids of the Krebs cycle are needed in catalytic quantities for the oxidation of carbohydrates. The studies of carbon dioxide support the hypothesis that the rise in the rate of absorption of oxygen by lemons under high partial pressures of carbon dioxide might be regulated by the formation of these acids.

The fact that malonate was found to be present and labelled during a prolonged exposure to $^{14}CO_2$ suggests that another mechanism may be involved. Figure 7 is a reproduction of an actual chromatogram showing evidence that malonate as well as citrate was found in some of the fractions. Unequivocal proof that the product was indeed malonic acid was obtained by independent chemical means.

Storage of lemons in atmospheres containing 5 or 10 per cent of carbon dioxide tends to suppress chlorophyll destruction (figure 6). All lemons were dark green at the beginning of the experiment, but, for any particular level of oxygen, the samples in atmospheres low in carbon dioxide coloured more rapidly than the ones under higher concentrations. Figure 6 also shows the action of ethylene in accelerating the decomposition of chlorophyll.

Ethylene has played an important role in fruit physiology ever since it was identified as the active agent in the combustion products to which green lemons were exposed. Minute quantities of this gas, of the order of 1 p.p.m., induce ripening, stimulate exchange of the respiratory gases, and hasten the onset of the climacteric rise. It should be noted that the two classes of fruits, the climacteric and the nonclimacteric, differ in their response to ethylene (figure 12). In the climacteric group ethylene is effective only if applied during the preclimacteric phase, and it acts only to shift the time axis, without altering the shape of the respiratory curve and without causing changes in the major constituents that would not occur otherwise. These responses are induced by concentrations of 0·1–1000 p.p.m. In the nonclimacteric group, respiration can be stimulated at any time during the life of the detached fruit. Within a wide range of concentration, the more ethylene is added to the air, the more pronounced the stimulation. The effects of ethylene were simulated also by volatile products of ripe fruit; those were found to induce ripening in green fruit. Similar action was brought about by emanations of fungi such as *Penicillium digitatum* and *Alternaria citri*. Since ethylene was identified as the active component in the volatile products of certain

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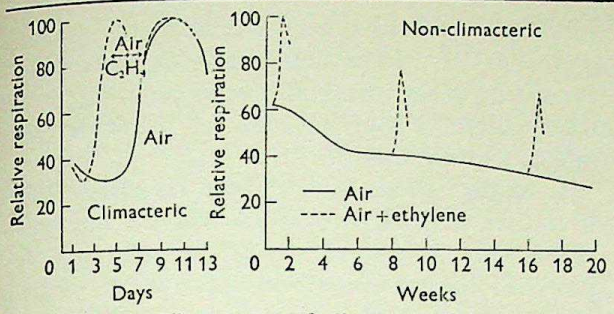

FIGURE 12 – *The response of climacteric and non-climacteric fruits to ethylene.*

fruits and fungi, intense interest developed in this hydrocarbon as a possible hormone causing ripening.

BIOGENESIS OF PARTICULAR CONSTITUENTS OF FRUIT

Ethylene. With the methods available until recently, it was found that the first detectable quantities of ethylene appeared at, or shortly after, the climacteric rise in respiration. The rates of ethylene evolution bore no direct relation to the rates of absorption of oxygen or evolution of carbon dioxide (figure 13). The hypothesis was, therefore, advanced that ethylene was a product of the ripening process rather than the causal agent. A re-examination of this problem has been undertaken in recent years, using gas chromatography, which measures smaller quantities. It was again established that the appearance of ethylene is an aerobic process, though a precursor may be formed anaerobically. Alcohol, which is a product of the fermentative process, was not converted into ethylene. This was established by injecting small quantities of radioactive ethanol into the vascular system of the cherimoya fruit at a time of intense ethylene production. The respiratory carbon dioxide was labelled at once while the ethylene was not. Under the same conditions, label from glucose did appear as ethylene though the specific activity was much higher in respiratory carbon dioxide. This was also the case when acetate, fumarate, formate, or pyruvate were injected. The available evidence favours the viewpoint that the precursors of ethylene are not intermediates in the major pathways of metabolism.

Anthocyanins. Most of the red pigments that enhance the colour in fruits are anthocyanins. The particular pigment found in apple skin is idaein (cyanidin-3-galactoside). It has long been known that apples exposed to sunlight develop more red colour than fruit held in the dark. H. W. Siegelman and S. B. Hendricks demonstrated this requirement for light under laboratory conditions (figure 4) and examined the action of light on anthocyanin synthesis with the aid of a high-intensity spectroscope [6]. They exposed segments of apple skin suspended in sucrose solution to light, using wavelengths from a wide region of the spectrum and they observed maximum anthocyanin production in those sections exposed to wavelengths between 6200 and 6900 Å. They proposed a scheme for anthocyanin formation that involved active acetate; two radiation steps led to the cyclization of a polyacetate compound into the phloroglucinol ring of anthocyanin. The same photoreceptor appears to be involved in both radiation steps. The action spectrum suggested that the photoreceptor was an acyl co-enzyme A dehydrogenase. The interesting point here is the contribution that the work with apple-skin tissue has made to the general problem of the effect of light on photomorphogenic processes. Red light stimulates germination in many seeds, and prevents flowering and reduces stem elongation in some plants. In each of these cases, exposure to infrared radiation reverses the action of red light.

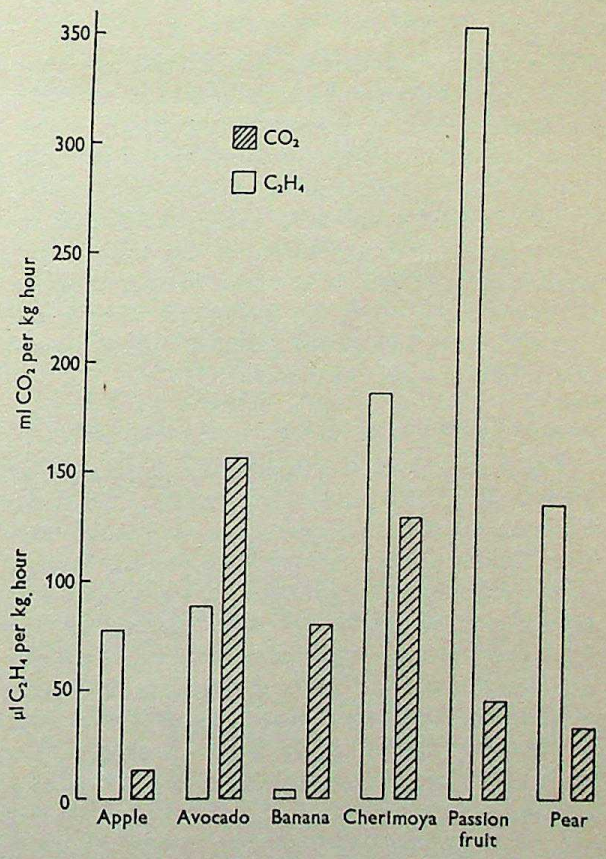

FIGURE 13 – *Ethylene production in relation to respiratory carbon dioxide evolution.*

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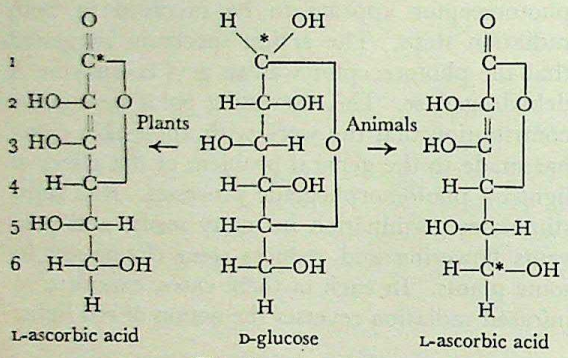
Conceivably other morphogenic responses of plants are controlled by the same mechanism.

Ascorbic acid biosynthesis. The synthesis of ascorbic acid (vitamin C) provides an interesting problem in comparative biochemistry. Tracer studies with D-glucose-1-^{14}C revealed [4] that ripening strawberries convert this sugar to L-ascorbic acid over a metabolic path that conserves all six carbons of glucose and results in an ascorbic acid molecule whose chain sequence is the same as that of the parent glucose. In the rat, on the other hand, the conversion of D-glucose to L-ascorbic acid results in an inverted carbon chain sequence.



C* radioactive carbon

The intermediate substances in the plant synthesis are unknown, although hexose phosphates are known to be involved in the initial steps. Strangely, when specifically labelled D-glucuronic acid, a known intermediate in the synthesis of ascorbic acid by animals, was administered to ripening strawberries, inversion did take place. Conceivably, plants make ascorbic acid in more than one way. It is an important nutritional factor for man but its role, if any, in fruit metabolism has not been elucidated.

RESPIRATORY EVENTS IN TISSUE DISCS

The experiments with intact fruit have furnished a great deal of valuable information, but have left many problems unanswered. It became evident that in order to investigate the mechanism responsible for the regulation of respiration and ripening, it would be necessary to separate the fruit cell from the influences of possible oxygen deficiency, carbon dioxide excess, and the action of volatile substances that may accumulate under a relatively impermeable skin. A slice of the flesh of an avocado or apple, or of the rind of a lemon or orange, suspended in a buffer of pH 5–7 takes up oxygen steadily during a period of one to several hours. The respiration is endogenous, which means that it takes place at the expense of substrates, such as sugars, stored in the cells. A great deal of information can be deduced about the nature of the metabolic events occurring in the tissue slice by the addition of inhibitors and labelled substances.

Cyanide, azide, and carbon monoxide have been used to block cytochrome oxidase, the terminal enzyme in the transfer of electrons from the material that is respired, the substrate, to oxygen. By the use of these inhibitors, it was shown that both oranges and lemons are sensitive to heavy-metal inhibitors, but more so in the immature than in the mature fruit. At all times a considerable fraction of the respiration is cyanide-resistant, implying the coexistence of two or more respiratory pathways.

Evidence from the use of inhibitors has to be confirmed by spectroscopic studies. The respiratory enzymes have characteristic absorption spectra for the oxidized and reduced forms, and the rate of change from one to the other form in the presence of suitable substrates and cofactors is an indication of the activities of particular enzymes under consideration. This work was done with avocado tissue, but with preparations obtained by breaking up the cells rather than with fruit discs. It will be discussed later.

The work with slices of avocado tissue furnished valuable information about respiratory energetics. It has been well established that the energy for the various activities of the cell, such as protein synthesis, ion accumulation against a concentration gradient, and growth is derived ultimately from the hydrolysis of 'high energy' phosphate compounds. The key role is played by adenosine triphosphate, ATP, which is formed as a result of oxidative or photosynthetic phosphorylation. In materials devoid of chloroplasts we are concerned only with phosphorylations associated with the oxidation of carbohydrates, organic acids, and fatty acids. When the loss of electrons (oxidation) is closely linked to ATP formation, it is referred to as coupled oxidation; in these circumstances the rate at which phosphorylation occurs limits the rate of oxidation. Uncoupling, the dissociation of respiration from phosphorylation, may be brought about by various physiological conditions and can be studied effectively by the use of 2,4-dinitrophenol, DNP, in appropriate concentrations.

It occurred to fruit physiologists that the increased oxidation associated with the climacteric in fruits might be the result of uncoupling, so that the rate of oxidation is no longer limited by the rate of phosphorylation. Slices of avocado

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were removed from fruit at the several stages of the climacteric and the effect of DNP on their oxidative activity studied. The results are given in table III. Clearly, the oxidation in the preclimacteric stage was stimulated by DNP, while at the peak of the climacteric it was not. Two explanations were possible for the increase in the rate of oxygen absorption by the intact fruit. (1) As a result of cellular activities at the onset and during the climacteric rise, ATP is utilized, and ADP, the phosphate acceptor required for coupled oxidation, is made available, or (2) a native uncoupling agent, similar in its action to DNP, is formed. If the second hypothesis holds rather than the first, phosphorylation of ripe avocado slices should be very low and should be unaffected by the addition of DNP. Experiments were undertaken, therefore, with labelled phosphate supplied to slices of the avocado tissue at various stages of ripening, and for various intervals of time, in the presence and absence of DNP. Immediately after treatment, the action of enzymes that split phosphate bonds was prevented by immersing the tissue in boiling methanol. After extraction, purification, and concentration, the various organic phosphates, as well as inorganic phosphates, were separated by two-dimensional chromatography and by chromatography in one direction followed by electrophoresis in the other. The spots on the chromatograms were located by X-ray radiography and by ultraviolet light. The spots were cut out and the radioactivity determined. The results clearly indicate that ^{32}P was incorporated into a number of esters including sugars, nucleotides, and three carbon acids. Contrary to expectation, the rate of incorporation of ^{32}P into these compounds was higher in slices at the climacteric peak than in preclimacteric slices. DNP caused decreased incorporation of ^{32}P into nucleotides and sugars in both preclimacteric and climacteric-peak slices. This showed that coupled phosphorylations were occurring in both stages. ^{32}P was found to be introduced into an unidentified compound that accounted for nearly 50 per cent of the activity in the esterified form, and incorporation into this compound was not inhibited by DNP in the post-climacteric stage. These data suggest that coupled phosphorylations do occur throughout the ripening process and that the turnover rate of the phosphorylative mechanism is increased. Interestingly, the phosphorylative mechanism appears to become more active, instead of 'wearing out', as the fruit enters senescence.

TABLE III

Dinitrophenol effect in relation to ripening

	Dinitrophenol concentration	Microlitres O_2/gram dry weight hour	
		Pre-climacteric	Climacteric Peak
Whole fruit ..	—	133	685
Tissue slices	0	640	630
	10^{-5} M	990	670
	10^{-4}M	1230	610

THE ROLE OF MITOCHONDRIA IN FRUIT METABOLISM

The mitochondrion is the energy generator of all cells, though in green plants chloroplasts are also a source of energy, derived from sunlight, for cellular reactions. Mitochondria may be spherical or rod-shaped, and range in size from 0·1 to 5 microns; they have been isolated from various types of cells. Although these particles are barely visible under the light microscope, electron microscopy has revealed their structure (figure 8). They consist of lipoprotein double exterior membranes enclosing a series of double-membraned lamellae. The outer membrane can stretch and swell enormously in media with low osmotic pressures. In order to isolate mitochondria capable of carrying out metabolic activities, the preparative procedure must be carefully controlled.

Considerable success was experienced in isolating active mitochondria from the avocado fruit [2], and preparations capable of oxidizing some Krebs-cycle acids have been obtained recently from other fruits. Temperatures close to 0°C, relatively high osmotic pressure of the medium,

TABLE IV

Oxidative activity shown by mitochondria of several fruits (microlitres O_2 per mg protein nitrogen per hour)

Fruit	Substrate				
	Succinate	α-Ketoglutarate	Malate	Citrate	Pyruvate
Apple ..	276	76	133	86	—
Avocado ..	455	211	252	284	334
Banana ..	235	85	134	151	—
Papaya ..	869	361	484	373	—
Pepper fruit	254	94	167	195	—

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pH close to neutrality, and low speed of blending are critical requirements in the preparation of active cytoplasmic particles from the avocado mesocarp. In some fruits, the presence of protein-inactivating substances, such as tannins, interferes with successful isolation. This condition has been counteracted in study of the banana by the addition of rather large amounts of an inert protein to the isolation medium; this lowers the concentration of the inactivator by reacting with it. Following homogenization, the unbroken cells and large fragments, such as cell wall material, plastids, and nuclei, are separated by a low centrifugal force. Centrifugation using forces up to 15 000 *g* brings down nearly the entire mitochondrial fraction. Metabolic activity is then determined by use of the Warburg apparatus, oxygen electrode, pH titrimeter, or spectrophotometer.

The ability of mitochondria from several fruits to oxidize Krebs cycle acids is shown in table IV. Thus far the avocado is the only fruit that yielded particles capable of oxidizing pyruvic acid, the link between carbohydrate metabolism and the Krebs cycle. Catalytic quantities of malate were required for the oxidation of pyruvate to citrate. Evidence that the tricarboxylic-acid pathway (Krebs cycle) operates in avocado mitochondria was supplied by the high rates of oxidation of key acids of the cycle, by inhibitor studies using malonate and arsenite, and by isolation of products of specific reactions. It was also shown that avocado particles possess a mechanism for the oxidation of reduced diphosphopyridine nucleotide (DPN), the cofactor of dehydrogenases, for oxidizing and reducing cytochrome c and for passing electrons to cytochrome oxidase, a widely occurring terminal enzyme in respiration.

The relationship of mitochondrial activity to ripening was demonstrated in studies involving oxidative phosphorylation. As a result of the oxidation of acids of the Krebs cycle, inorganic phosphate is converted into organic form, notably into ATP. The ratio of micromoles of phosphate esterified to micro-atoms of oxygen utilized is a measure of phosphorylative efficiency. This ratio tends to increase with the course of fruit ripening;

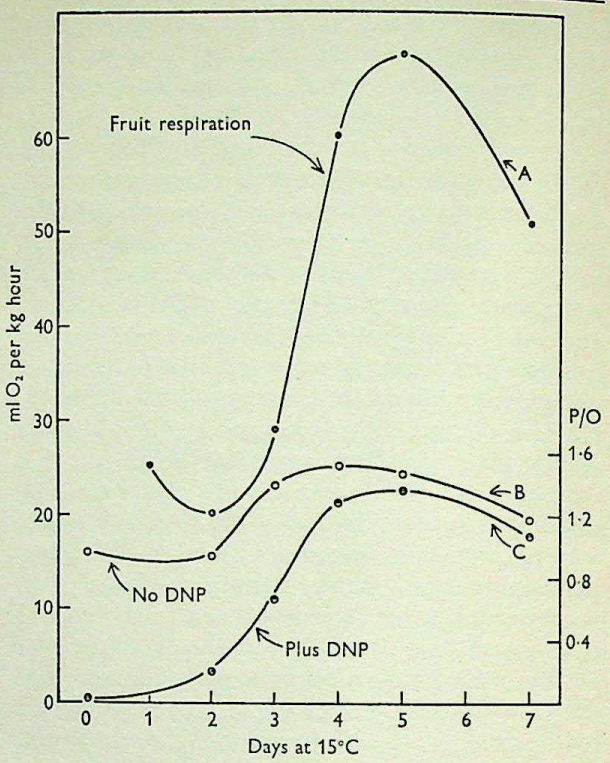

FIGURE 14 – *Effect of 2,4-dinitrophenol on the P/O ratio of mitochondria isolated at various stages of the climacteric. Curve A: Respiration (oxygen uptake) of a typical intact fruit. Curves B and C: P/O ratios of mitochondria isolated from fruits at the various stages of the climacteric with pyruvate as the substrate.*

the increase is frequently due to lower oxidation rates as well as to increased phosphorylation. Mitochondria from preclimacteric fruit are nearly completely uncoupled by DNP, while these particles from fruit at the climacteric peak are affected only slightly by the uncoupler (figure 14). This behaviour of mitochondria and the phosphate pattern obtained from the tissue-slice studies suggests that incorporation of phosphate into certain specific substances may be the key answer to the metabolic changes in the ripening fruit. The precise meaning of DNP-resistant phosphorylation requires further elucidation. The degree of coupling of oxidation with phosphorylation is a problem of general biological importance and a field of active research by both plant and animal biochemists.

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Hydrocarbon derivatives of transition metals

P. L. PAUSON

This article reviews some of the recent investigations of organometallic derivatives of transition metals, with emphasis on the first-row transition metals. Apart from their intrinsic interest, some of these novel compounds have added considerably to our understanding of aromaticity, by which the organic chemist means a tendency to undergo electrophilic substitution reactions. Others have helped us to understand the mechanisms of reactions in which they are involved as homogeneous catalysts. Although these new substances have as yet found few practical applications, their possibilities appear to be very considerable.

Transition metals can be bonded directly to carbon atoms of a wide variety of hydrocarbon groupings. Recognition of this fact, which is contrary to views widely held little more than ten years ago, is one of the more exciting recent developments in chemistry [1]. Then, only the metal carbides, the cyanides, and the carbonyls and closely related isonitrile complexes were definitely known to contain direct links between these metals and carbon atoms.

Ever since Frankland's discovery of the zinc alkyls, interest in organometallic compounds has been strong, but for a long time all attempts to obtain transition-metal derivatives failed. Methods like the metal-exchange reaction, so successful in other cases, for example:

$$4RMgCl + SnCl_4 \rightarrow R_4Sn + 4MgCl_2$$

where R is an alkyl group, did not yield isolable compounds when applied to the halides of such metals as manganese, iron, or cobalt. However, since such halides are reduced by Grignard reagents it was reasonable to postulate intermediate formation of alkyls RMX or R_2M, where M is the metal. Recent work has shown that at very low temperatures such alkyl derivatives can be prepared from Mn^{II}, Cr^{III}, and Ti^{IV}. Indirect evidence has been obtained for the existence of analogous compounds of Ni^{II} and Co^{II}. The earlier failures are explained by the fact that all these compounds decompose below room temperature, probably by a simple splitting off of the alkyl groups to give dimeric hydrocarbons and the free metal:

$$R_2M \rightarrow R{-}R + M$$

If we look more closely at the stable alkyls formed from other metals [2], it will be seen that in type these resemble the corresponding halides. Thus, the strongly electropositive alkali metals form essentially ionic alkyls (for example $CH_3^- K^+$), while the tetra-alkyls of tin (for example $(CH_3)_4Sn$) resemble the covalent halides, such as $SnCl_4$, which exist as discrete molecules at least in the vapour phase. Most transition metals do not form halides of these types. Thus cobaltous chloride ($CoCl_2$) is commonly met in the hydrated form $[Co(H_2O)_6]Cl_2$ in which the metal is bound directly to six oxygen atoms. In the anhydrous compound it attains a similarly high coordination number by forming a polymeric structure.

It would be reasonable, therefore, to expect that stable organic compounds of these elements must also involve a suitably high coordination number, coupled with a suitable electronic configuration. Empirically, it is found that by far the most stable organic compounds of transition metals are those in which the metal can be formulated as having attained the electronic configuration of the next inert gas. The valency or oxidation state of the metal is apparently of only secondary importance.

W. Reppe, in his wartime work on reactions of unsaturated hydrocarbons and alcohols with carbon monoxide in the presence of cobalt carbonyl, postulated the formation of alkyl cobalt

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carbonyls, $RCo(CO)_4$, as probable intermediates [3]. For example, he believed that methylcobalt tetracarbonyl, $CH_3Co(CO)_4$ was involved in the transformation of methyl alcohol to acetic acid according to the equation

$$CH_3OH + CO \xrightarrow{Co_2(CO)_8} CH_3COOH$$

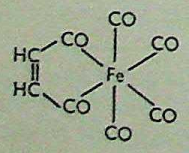
However, no such alkyl compounds were known, and the manganese compounds, $RMn(CO)_5$, the first stable alkyls of similar type, were first described only in 1957. They are prepared by reduction of dimanganese decacarbonyl with sodium amalgam, followed by treatment of the sodium derivative with the appropriate halide:

$$Mn_2(CO)_{10} + 2Na \rightarrow 2NaMn(CO)_5 \xrightarrow{RX} NaX + RMn(CO)_5$$

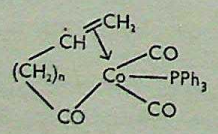
An alkyl halide or an acid chloride may be used; for example, methyl iodide yields methylmanganese pentacarbonyl (I). Similarly, acetyl chloride yields acetylmanganese pentacarbonyl (II). Such alkyl and acyl derivatives are themselves interconvertible, for example:

$$\underset{\text{I}}{CH_3Mn(CO)_5} + CO \underset{\text{heat}}{\rightleftarrows} \underset{\text{II}}{CH_3COMn(CO)_5}$$

Significantly, these very stable manganese compounds are hexacoordinate and the manganese atom (atomic number 25) obtains one electron by forming the covalent bond with the alkyl (or acyl) group and an additional ten electrons from the five coordinate bonds with the carbonyl groups. This gives the manganese atom an effective atomic number of 36, equal to that of krypton. The electron-withdrawing effect of the carbonyl group makes the acyl derivatives more stable than the alkyl derivatives. The same effect is met in enhanced degree when we compare the hydrocarbon complexes with their perfluoro analogues. Thus both $C_nF_{2n+1}Co(CO)_4$ and $C_nF_{2n+1}Mn(CO)_5$ are considerably more stable than their alkyl analogues. Even iron gives similar fluorocarbon complexes, whereas analogous alkyls are not known. The chelated di-acyl compound (III) has been obtained by an indirect route, starting from iron carbonyl and acetylene.



III



IV

The additional stabilization to be obtained from bifunctional groups that can be linked to the metal at both ends to form a ring (chelate groups) is, of course, well known for the more conventional coordination compounds of the transition metals. It is hardly surprising to meet it again here, especially in compounds, like III, that are typical octahedral complexes.

In the cobalt series, it has also been shown that further stabilization results on replacement of one carbonyl by a phosphine group. Thus the reaction $RCo(CO)_4 + PPh_3 \rightarrow RCOCo(CO)_3PPh_3$ readily affords a product more stable than the analogous acyl cobalt tetracarbonyl $RCOCo(CO)_4$. With acyl groups of the type $CH_2{=}CH(CH_2)_nCO$— products of type IV have been isolated. Here, the carbon-carbon double bond has displaced one CO group; we then have a simple olefine complex at the same time as an acyl derivative, both no doubt stabilized by chelation. Few other simple-olefine complexes have yet been isolated. The most notable example is

$$\underset{\text{V}}{\left[Mn(CO)_5 \leftarrow \begin{matrix} CH_2 \\ \| \\ CH_2 \end{matrix}\right]^+}$$

obtained from bromomanganese pentacarbonyl ($BrMn(CO)_5$) and ethylene in the presence of aluminium bromide. The ethylene here forms a type of bond that was first recognized in platinum complexes, which have been known much longer than the above examples involving metals of the first transition series.

The double bond in ethylene itself may be described, in molecular orbital terminology, as consisting of two parts, of different types. The first, or σ, bond is a normal single bond, as in ethane. If we consider a molecule CH_2—CH_2 formed with just such a bond (that is, with each carbon atom forming three sp^2 hybrid bonds) we arrive at the picture (A) in which each carbon has one more electron in a p-orbital (shaded). When these electrons are paired (to give us the ethylene molecule $CH_2{=}CH_2$) they will lie in a new hybrid orbital whose approximate shape is shown in B. This type of bond is referred to as a π-bond. The electrons in this π-orbital can be donated to a metal atom to form a normal co-ordinate bond. The approximate overlap of the π-orbital of ethylene with a suitable d-orbital of the metal is shown in C. At the same time, electrical neutrality is restored by back-donation of electrons from a filled d-orbital of the metal into

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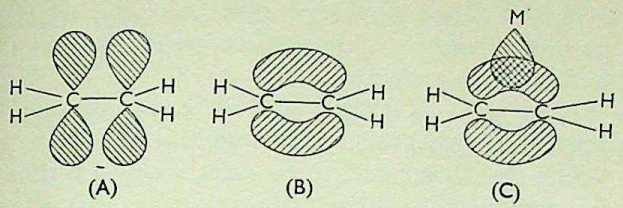

the antibonding π-orbital of the ethylene; this is not shown. A compound formed in this way is commonly termed a π-complex between the hydrocarbon and the metal atom.

Compounds will next be considered in which more extended π-electron systems are involved. The product formed from allyl bromide and sodiummanganese pentacarbonyl depends on the temperature:

$$CH_2{=}CH{-}CH_2Br{-}$$
$$+NaMn(CO)_5 \xrightarrow{20^\circ} CH_2{=}CH{-}CH_2Mn(CO)_5$$
$$\xrightarrow{80^\circ} \begin{matrix} CH_2 \\ CH \\ CH_2 \end{matrix} \!\!\!\

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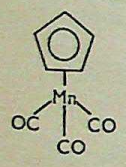
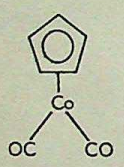
As an example of a 5-electron donor grouping the cyclopentadienyl radical C_5H_5 stands supreme. Its sandwich complexes with transition metals were the first of these π-complexes whose structure was elucidated. The outstanding example, ferrocene, $(C_5H_5)_2Fe$ (XI) and its simple analogues formed one of the subjects of a review in ENDEAVOUR in 1955 [5].

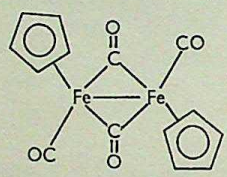
Since then, ferrocene itself has been studied very extensively, especially from the point of view of its behaviour as a novel aromatic system; to the dicyclopentadienyl derivatives of other metals then listed, a wide range of monocyclopentadienyl derivatives can now be added. Among the latter, the carbonyls are the best known and include such compounds as the monomeric manganese (XII) and cobalt (XIII) derivatives and the dimeric iron compound (XIV). However, carbon monoxide is not a unique ligand in such compounds, and other ligands can give rise to equally stable derivatives, as illustrated by the nitrosyl complex of nickel (XV), the trichloride of titanium (XVI),



XII

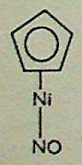


XIII

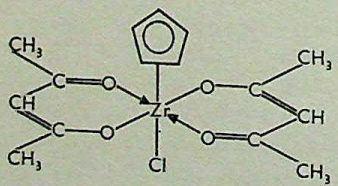


XIV

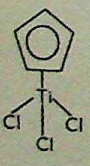
and the acetylacetonate complex of zirconium (XVII). The carbonyl complex of cobalt (XIII) reacts readily with dienes, conjugated or unconjugated, to give the mixed cyclopentadienyl diene complexes (for example, XVIII $n=2$; $m=0$; or $n=m=2$).



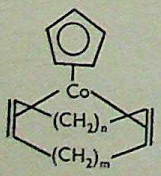
XV



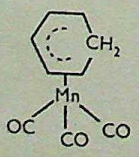
XVII



XVI



XVIII



XIX

Recently, these complexes have been supplemented by the isolation of cyclohexadienyl and cycloheptadienyl derivatives, for example the cyclohexadienyl analogue (XIX) of (XII), which provide examples of alternative 5-electron donor hydrocarbon groupings.

In 1955, L. Onsager and H. Zeiss suggested that F. Hein's polyphenylchromium derivatives, which had long puzzled chemists, were in reality π-complexes of benzene and diphenyl having such structures as XX, where R is a phenyl group. Clear proof was provided by the preparation of the parent compound, dibenzenechromium (XX, R = H), by E. O. Fischer and W. Hafner. Other complexes of aromatic hydrocarbons soon followed, including the benzenechromium tricarbonyl type (XXI). Poly- and even heterocyclic aromatics (thiophene) form compounds analogous to (XXI), and so do conjugated trienes. Of the latter, cycloheptatriene forms with chromium the complex (XXII), which loses a hydride ion to give the cationic tricarbonyltropylium-chromium (XXIII). When vanadium carbonyl became available recently, the synthesis of the neutral tropylium-vanadium tricarbonyl (XXIV), isoelectronic with the chromium cation (XXIII), became possible. These two substances contain the C_7H_7 group; this may be considered to act as a 7-electron donor, thus completing the series so far as it is known today.

The complexes listed above have been chosen as illustrative of the main types. They are known to undergo many interesting transformations to other complexes or even interconversions. Some have thrown new light on aromaticity and others on some processes involving homogeneous catalysis. It is convenient to describe these aspects in separate sections.

TRANSFORMATIONS

The dicyclopentadienyl-di-iron tetracarbonyl (XIV) provides characteristic examples of many reactions. It is converted on pyrolysis to ferrocene (XI), with migration of one of the rings from one iron atom to the other. Simple replacement of carbonyl groups has been effected with dimethyl disulphide which produces the bridged sulphide (XXV).

Sodium amalgam reduces the tetracarbonyl (XIV) to the sodium salt (XXVI) which reacts readily with alkyl and acyl halides to give stable derivatives (XXVII; R = alkyl or acyl). The halides (XXVIII; X = Cl, Br, or I), formed on oxidation of the binuclear carbonyl (XIV) with the appropriate halogen, have been used to obtain mixed bimetallic derivatives, e.g.

$$C_5H_5Fe(CO)_2—Mn(CO)_5$$

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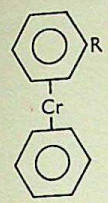
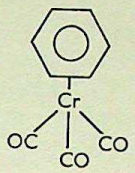
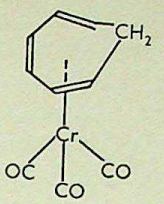


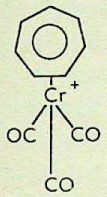
XX



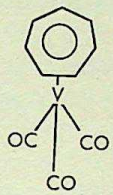
XXI



XXII



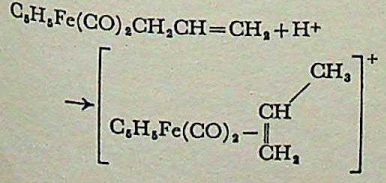
XXIII



XXIV

by treatment with sodiummanganese pentacarbonyl, or $C_5H_5Fe(CO)_2$—$Mo(CO)_3C_5H_5$ on treatment with sodiumcyclopentadienylmolybdenum tricarbonyl, $C_5H_5Mo(CO)_3Na$. Unlike the iron complex (XIV), both these mixed compounds are believed to be held together only by a metal-metal bond. As in the binuclear manganese and molybdenum complexes $Mn_2(CO)_{10}$ and $(C_5H_5)_2Mo_2(CO)_6$ to which they are related, this probably allows the metals to reach an optimum coordination number.

In the compounds of type XXVIII, the halogen atom, X, can be replaced by other anions, for example, cyanide, or be displaced out of the coordination sphere by neutral ligands to give cationic products. Thus triphenyl phosphine gives salts of the ion $[C_5H_5Fe(CO)_2PPh_3]^+$ and in the presence of aluminium bromide, carbon monoxide, or ethylene reacts to give the related ions (XXIX) and (XXX; R = H) respectively. The propylene complex (XXX; R = CH_3) has been obtained similarly, and also by treating the σ-allyl derivative (XXVII; R = $-CH_2CH{=}CH_2$) with acid. A rearrangement of the allyl group occurs in the reaction:

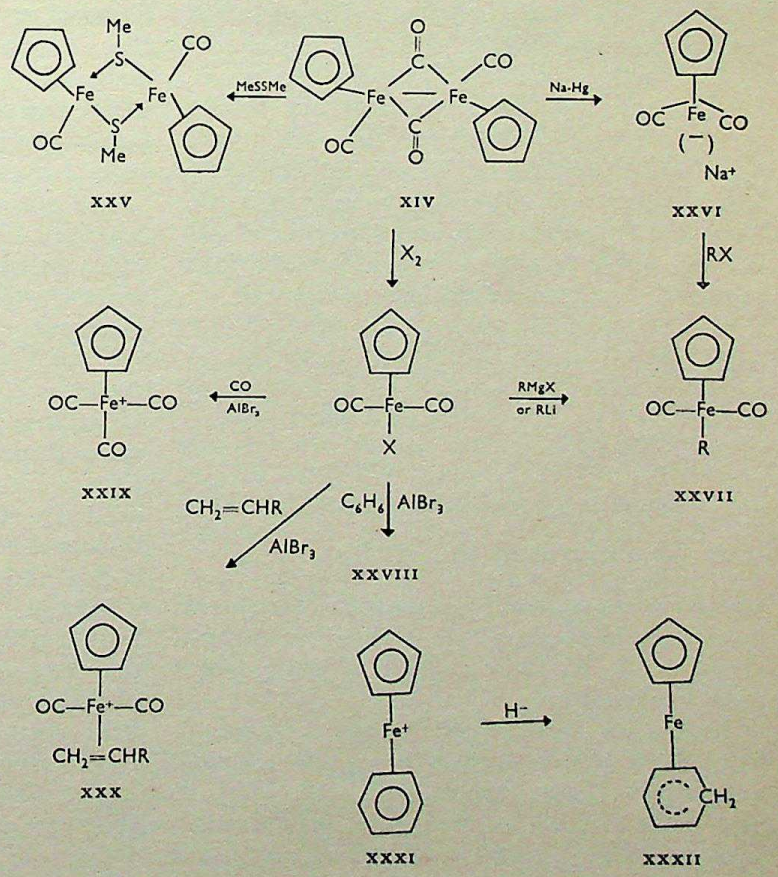


Treatment of the halides (XXVIII) with benzene in the presence of Friedel-Crafts catalysts gives the cyclopentadiene-iron-benzene cation (XXXI). This will add anions to the six-membered ring, notably hydride ions (as boro- or aluminohydride), to give cyclohexadienyl-cyclopentadienyl-iron (XXXII), a close analogue of ferrocene (XI).

A variety of groups have been used to displace one cyclopentadiene ring from dicyclopentadienyl nickel $(C_5H_5)_2Ni$ (XXXIII). The exceptional ease with which one of the two rings can be displaced in this case may be attributed to the fact that this complex has two electrons in excess of the inert gas configuration. Thus it reacts with nitric oxide to give the nitrosyl (XV); with iron and nickel carbonyls to yield the binuclear compounds (XXXIV) and (XXXV); and with acetylenes to give the acetylene-bridged complexes (XXXVI) also obtainable from the dicarbonyl (XXXV).

AROMATICITY

The organic chemist's chief criterion of aromaticity is the tendency to undergo electrophilic substitution reactions. As was first shown by Hückel, molecular orbital theory predicts that



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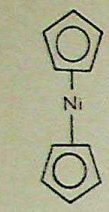
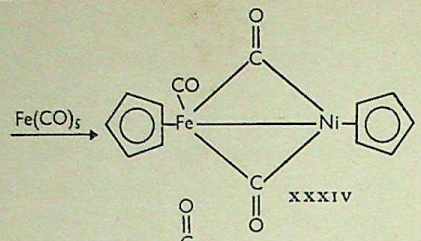
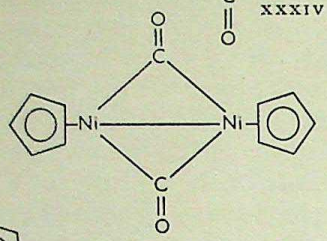


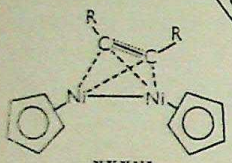
XXXIII



XXXIV

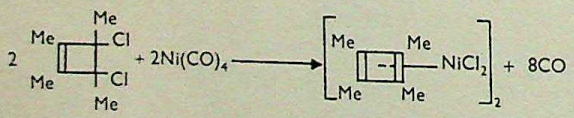


XXXV

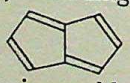


XXXVI

only those monocyclic systems in which the number of π-electrons is $4n+2$ will be aromatic. In practice, this restricts aromaticity to 6π-electron systems among the commonly available homo- and heterocyclic systems with one ring. Both cyclopentadienide ions, $C_5H_5^-$, and tropylium ions, $C_7H_7^+$, are aromatic systems of this kind. But only with the discovery of their transition-metal complexes have these odd-membered rings become available in the form of neutral molecules whose aromaticity can be tested and exploited by the characteristic substitution reactions so familiar with aromatics derived from benzene. Moreover, Hückel's rule is not, of course, applicable to such metal complexes and it is found that even neutral $4n$ π-electron systems can form metal complexes which are certainly stable and probably aromatic. Thus the tetramethylcyclobutadiene-nickel dichloride dimer (XXXVII) is formed from dichlorotetramethylcyclobutene and nickel carbonyl:



XXXVII

Probably other $4n$ π-electron systems, including such bicyclic systems as pentalene,  which are non-aromatic and unstable or incapable of existing in the free state, will, like cyclobutadiene, be found to be capable of giving stable metallic complexes.

Several of the cyclopentadienyl compounds have been shown to possess typical aromatic properties. They include ferrocene (XI); its ruthenium and osmium analogues; and the monocyclopentadienyl metal carbonyls of manganese (XII) and vanadium, $C_5H_5V(CO)_4$. The manganese complex (XII) has been shown to undergo both sulphonation and Friedel-Crafts acylation and alkylation. A considerable range of derivatives has been prepared by these processes and by further modification of the substituents so introduced, but because it is the most readily available example, ferrocene has received by far the greatest attention [6]. The ease with which it undergoes substitution, being comparable in this respect with phenol, and the stability of most of its derivatives make it particularly suitable for study. The chief limitation is imposed by its ability to be oxidized to the ferricinium ion, $(C_5H_5)_2Fe^+$, which is no longer reactive towards electrophilic agents. As would be expected, electron-donating substituents facilitate this oxidation; conversely electron-withdrawing groups make it more difficult. Thus the hydroxy- and amino-ferrocenes are sensitive even to air, while acyl derivatives are stable. As the manganese tricarbonyl complex (XII) is also sensitive to oxidation, such substitutions as nitration or halogenation are impossible in both series of compounds and special methods have to be employed to introduce certain substituents. It is possible to illustrate this here by only a few examples. For example, it is possible to obtain ferrocene-carboxylic acid, $C_5H_5FeC_5H_4CO_2H$, by Friedel-Crafts reaction of ferrocene and acetyl chloride and hypochlorite oxidation of the resultant acetyl ferrocene.

The nitro- and halogeno- derivatives of ferrocene are obtainable by indirect methods in which metallation is the first step. This is conveniently effected with butyllithium:

$$C_4H_9Li + C_{10}H_{10}Fe \rightarrow C_4H_{10} + C_5H_5FeC_5H_4Li$$

XXXVIII

In practice, the reaction does not stop at the monosubstituted derivative (XXXVIII) shown, but gives a mixture of this and the symmetrical dilithium derivative, $Fe(C_5H_4Li)_2$. When the lithium-derivative (XXXVIII) is allowed to react with propyl nitrate, nitroferrocene, $C_5H_5FeC_5H_4NO_2$, is formed. When the same lithium-derivative is treated with butyl borate, $B(OC_4H_9)_3$, the ferrocenylboronic acid $C_5H_5FeC_5H_4B(OH)_2$ is readily obtained. It is accompanied by the diboronic acid, $Fe[C_5H_4B(OH)_2]_2$, formed from the dilithium derivative. Both these acids react with copper halides to give the mono- and di-halogenated ferrocenes, and with copper acetate, giving the acetates $C_5H_5FeC_5H_4OCOCH_3$ and

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$Fe(C_5H_4OCOCH_3)_2$ respectively, and hence provide routes to the hydroxyferrocenes.

Two examples must suffice to show the high reactivity of ferrocene. Firstly, it will condense with methylene-bisdimethylamine, $CH_2(NMe_2)_2$, with only acetic acid as a catalyst (and solvent), to give the amine $C_5H_5FeC_5H_4CH_2NMe_2$. Secondly, the aldehyde $C_5H_5FeC_5H_4CHO$ is formed when the parent compound is treated with N-formyl-N-methylaniline and phosphorus oxychloride (Vilsmeier reaction). Replacement of iron by ruthenium or osmium causes a progressive reduction in this reactivity. It has been suggested that this is due to stronger bonding between the rings and these metals, leaving a lower electron density on the rings. Cyclopentadienylmanganese tricarbonyl (XII) also is much less reactive than ferrocene, being much more closely comparable in this respect to benzene itself. We can look upon the $Mn(CO)_3$ group as having a typical electron-withdrawing effect on the ring, similar to that of NO_2 when attached to benzene.

It is also relevant to mention here the resistance of ferrocene to addition reactions. Thus catalytic hydrogenation to iron and cyclopentane requires extreme conditions (H_2/Raney nickel catalyst at about 350°) and the phenyl group in, for example phenylferrocene, $C_5H_5FeC_5H_4C_6H_5$, may be smoothly reduced (H_2/Pt at room temperature) without affecting the ferrocene nucleus. The latter is, however, broken down by very strong electron donors, such as lithium in ethylamine:

$$(C_5H_5)_2Fe + 2Li \xrightarrow{EtNH_2} 2C_5H_5Li + Fe$$

METAL CARBONYL CATALYSIS

Much of the stimulus for work on the metal carbonyl derivatives described above came from Reppe's work on the hydroformylation reactions (oxo reactions) and other reactions of olefines and acetylenes with carbon monoxide in the presence of nickel, cobalt, or iron carbonyls. From the latter, Reppe isolated several complexes as by-products. Others have subsequently been obtained by related processes. The more stable of these complexes are unlikely to be of significance except as by-products, while the more reactive complexes which are the intermediates in such 'catalytic' processes may be either not isolable or obtainable only when prepared by alternative methods under much milder conditions. Nevertheless, the evidence that has accumulated in recent years enables us to have a much clearer understanding of these reactions. Thus although it is not yet possible to advance complete mechanisms, at least highly probable outlines can be suggested.

The cobalt-catalysed hydroformylation is known to involve cobalt hydrocarbonyl as the 'real catalyst' and is undoubtedly initiated by the replacement of one carbonyl by an olefine, probably in two steps, thus:

$$HCo(CO)_4 \rightleftharpoons CO + HCo(CO)_3$$

$$HCo(CO)_3 + CH_2{=}CHR \rightleftharpoons HCo(CO)_3{-}\begin{matrix}CH_2\\ \|\\ CHR\end{matrix}$$

This is followed by rearrangement of the olefine-π-complex to an alkyl which rapidly stabilizes itself by recombining with carbon monoxide:

$$2HCo(CO)_3{-}\begin{matrix}CH_2\\ \|\\ CHR\end{matrix} \rightleftharpoons RCH_2CH_2Co(CO)_3 + CH_3CHRCo(CO)_3$$

$$RCH_2CH_2Co(CO)_3 + CO \rightleftharpoons RCH_2CH_2Co(CO)_4$$

$$CH_3CHRCo(CO)_3 + CO \rightleftharpoons CH_3CHRCo(CO)_4$$

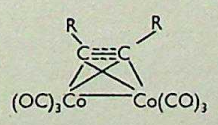
The initial release of CO can account for the known inhibiting effect of this gas. The reversibility at each stage can be shown to account for the varying isomer ratios in products derived from an unsymmetrical olefine $CH_2{=}CHR$, for, under different reaction conditions, the ratio may be controlled by either kinetic or thermodynamic factors. The two isomeric products reached by the above stages belong to the alkylcobalt tetracarbonyl type, $RCo(CO)_4$, isolable at low temperature when prepared from sodiumcobalt tetracarbonyl, $NaCo(CO)_4$, and alkyl halides. The succeeding steps can therefore be studied separately, and probably involve

(i) $RCo(CO)_4 \rightleftharpoons RCOCo(CO)_3$

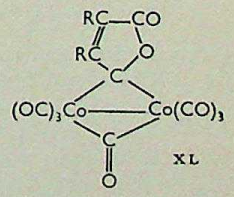
(ii) $RCOCo(CO)_3 + H_2 \rightleftharpoons RCOCoH_2(CO)_3$

(iii) $RCOCoH_2(CO)_3 \rightarrow RCH{=}O + HCo(CO)_3$

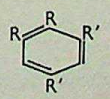
With acetylenes ($RC{\equiv}CR$) cobalt carbonyl reacts to give complexes of the type XXXIX. With carbon monoxide under pressure these react further to yield (XL).



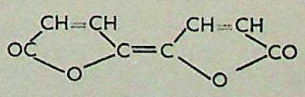
XXXIX



XL



XLII



XLI

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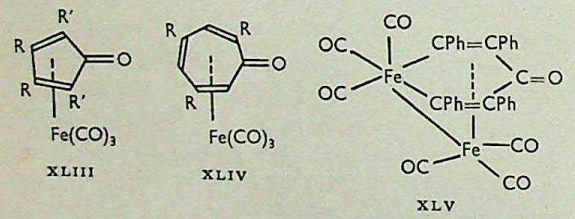
Acetylene has been shown to react with carbon monoxide and cobalt catalysts to give a variety of products, including the dilactone (XLI). As this is clearly derivable from XL (R = H), we have here an obvious outline of the route by which one of the more complex and unexpected products of such a reaction can be produced. The first of these complexes (XXXIX) is also a possible intermediate in the trimerization of acetylenes to benzene derivatives, using the mercury derivative, $Hg[Co(CO)_4]_2$, as catalyst. The intermediate (XXXIX) reacts not only with the same acetylene compound (RC≡CR) but also with different acetylenes such as R′C≡CH to give mixed benzene derivatives (XLII) including, for example, the novel and highly sterically hindered di- and tetra- tert-butyl derivatives (XLII, R = Bu^t R′ = H, namely R = R′ = Bu^t).

The variety and complexity of new structures that can result from acetylenes and metal carbonyls have been well illustrated by W. Hübel and his co-workers in their extensive studies of the reactions of phenylacetylene and tolane with carbonyls of iron. The products are numerous and highly complex. Among the simpler types are both the cyclopentadienone complexes (XLIII) and the tropone derivatives (XLIV). A complex



precursor of the latter has also been identified. Others include the substance XLV, which has been shown to decompose to tetraphenyl-1,4-benzoquinone on treatment with nitric acid. This sequence may well be related to the production of hydroquinones from acetylenes and iron carbonyls that has been described by Reppe.

These examples are but a few of the many that could be mentioned. They indicate that studies of such organometallic complexes help us to understand known processes in which the simple metal carbonyls act as homogeneous catalysts. But more important, they are a challenge to us to find new processes in which these new substances themselves are utilized either as catalysts, with perhaps novel properties or higher specificity, or as direct synthetic intermediates—as in the above-mentioned example of the preparation of 1,2-di-tert-butylbenzene derivatives, undoubtedly the first of many future applications.

Much effort has already been devoted to finding commercial uses for some of these compounds. Ferrocene itself was shown to inhibit carbon deposition and smoke formation when added to fuel oils, but so far this has not led to economically useful practical applications. Derivatives of ferrocene have been suggested as anti-anaemic agents and even as absorbers of ultra-violet light, to be used for the protection of future astronauts. A titanium analogue, dicyclopentadienyltitanium dichloride, $(C_5H_5)_2TiCl_2$, is one of the derivatives of this metal, which, together with aluminium compounds, promote the stereospecific polymerization of olefines, but to date it too has failed to find actual commercial use. Even the extent to which methylcyclopentadienylmanganese tricarbonyl, $CH_3C_5H_4Mn(CO)_3$, a homologue of (III), will find practical application seems to depend on the characteristics of the internal combustion engine of the future, although its effectiveness as an anti-knock agent and its synergism when applied in conjunction with tetraethyllead seem to have been amply demonstrated.

Thus the future holds promise of many further applications of the compounds described here; in the meantime a truly exciting new group of chemical compounds awaits fuller exploitation and examination.

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The mountain flora of the Malaysian tropics

C. G. G. J. VAN STEENIS

The tropical flora is very much richer than that of the temperate lands. In the mountains, the plants form zones of different species at different altitudes. These zones do not, however, correspond simply to variations in height and temperature, but can be shown to be the result of more complex ecological changes. This article discusses the zoning of the flora, and the reasons for it. It is illustrated with pictures of typical tropical mountain plants of Malaysia.

A visitor from the temperate zone arriving in the lowland tropics will meet almost everywhere the same kinds of cultivated ornamental plants, irrespective of whether he is in the Old or the New World—stately coconut palms, red and yellow hibiscus, fragrant frangipanis, luxuriant bougainvilleas smothered in red and purple, umbrella-shaped flame-trees displaying their gorgeous scarlet flowers. Such widely and commonly grown plants give the impression that the flora is the same throughout the tropics. The weeds near dwellings are likewise often the same in many tropical countries. Moreover, the native flora along the coast, on sandy beaches, in gloomy mangrove swamps, and in forests along tidal shores tends to have a similar aspect throughout the tropics. Hence in Linnaeus's time, when the tropical flora was mostly known from specimens gathered near ports and around other lowland towns, it was assumed to be much the same everywhere between the Tropics of Cancer and Capricorn and no richer than the temperate flora. As soon, however, as botanists investigated the untouched vegetation of the interior, they met, as James Wallace [6] did in Darien in 1698, 'regions of monstrous Plants, enough to confound all the methods of Botany ever hitherto thought upon'. It is now recognized that the tropical flora far exceeds the temperate flora in number of genera and species and that they differ greatly from country to country. Certain regions are very rich in plants with restricted areas, and this flora is now seen to represent the core of the morphological evolution and development of the flowering plants, the temperate flora consisting of outlying derivatives from this. Botanical teaching based on the temperate flora must necessarily be ill-balanced and inadequate. The overwhelming richness of the tropical flora still remains evident when, in ascending the hills and mountains of the interior, we find the tropical lowland flora gradually replaced by the equally rich and varied montane lauro-fagaceous forest teeming with epiphytic plants (plants that grow attached to the twigs or stem of another plant), mostly orchids and ferns and, in the cloud zone, mosses and liverworts. In yet higher zones the forest gradually takes on a more temperate character; its canopy is lower, its composition is less mixed, and in it there appear conifers and representatives of genera characteristic of temperate regions such as *Anemone*, *Valeriana*, *Viola*, *Ranunculus*, *Lonicera*, *Berberis*, and *Taxus*. Towards an altitude of 4000 m in the Malaysian region the woody vegetation becomes dwarfed, with an increasing number of grasses, sedges, and herbs in its glades, and at about 4100 m it is replaced by open alpine meadow. If the mountains are high enough, as for example, the Snowy Mounts of New Guinea, all these zones are fully developed. From 4700 m upwards eternal snow and glaciers cap the summits.

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Was born in 1901 and was educated at the University at Utrecht, from which he received a Ph.D. degree in 1927. From 1927 to 1946 he was a botanist in the Herbarium of the Botanic Gardens at Buitenzorg (Bogor) in Java. Since 1950 he has been director of the Foundation Flora Malesiana and general editor of the 'Flora Malesiana'; he is also an honorary professor of Tropical Botany at Amsterdam and Leyden Universities. He works mainly at the *Rijksherbarium*, Leyden. He has published over two hundred papers dealing with floristics and taxonomy of Holland and Malaysia, ecology and vegetation, nature preservation, specific delimitation, biography, bibliography, and plant geography.

ALTITUDINAL ZONATION

The altitudinal zonation just described matches almost exactly the zonation by latitude from the equator towards the poles; both pass from tropical, subtropical, and warm temperate to temperate

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and cold conditions and floristic types. By applying in the tropics O. Sendtner's method of floristic statistics for altitudinal ranges [1], used by him in 1854 in the Bavarian Alps, I have been able to show [2] that the mountain flora has rather abrupt altitudinal breaks in composition and may be divided into the following zones. There is a tropical zone up to 1000 m (with a colline subzone from 500 to 1000 m), a montane zone from 1000 to 2400 m (with a submontane subzone from 1000 to 1500 m), a subalpine zone from 2400 to 4100 m, and above that the alpine and nival zones to and above the snow line respectively. A similar statistical examination of the ranges of crops and other cultivated plants has revealed the significance of the 500, 1000, and 1500 m boundaries in the ever-wet tropics of Malaysia, where the villages and fields are not found above 2000 m. This is in contrast to the Andes and the Himalayas, where they ascent much higher.

Although details vary, depending on situation and climate, the trend in zonation is essentially the same throughout the tropics, whether one climbs the Andes, Kilimanjaro, the Himalayas, Mount Kinabalu, or the Papuan Snow Range. The problems connected with it confronted the early explorers of the tropical mountains, notably Alexander von Humboldt in the Andes in 1802 and F. Junghuhn in Java in 1836, and have been the subject of major studies by O. Stapf [3] and by myself [2]. On active volcanoes the normal climatic zonation is, of course, much disturbed.

The altitudinal zonation has two independent aspects, the one concerning its physiognomy, that is, its external appearance, the other its floristic composition. The habit of growth of the vegetation changes with ascending altitude from many-storeyed, very mixed, lofty lowland forest to a one-storeyed lower and thinner forest, and then to a scrub, which in turn gives way to alpine heath and grassland. Even low, isolated, steep peaks, such as Mount Salhutu in Amboina and Mount Ranai in the Natuna Islands, which reach only 1000 m, show this kind of change in physiognomy. Their summit or crest, the highest elevation in a wide expanse of sea, attracts clouds almost every day, and the zoning of their vegetation imitates that of the high mountains in a telescoped way: the mossy forest and scrub on the summit at 800–1000 m exactly matches the physiognomy of the subalpine vegetation on the high mountains at 2000–3500 m. From this resemblance it can be concluded that the physiognomy must be largely due to topography, exposure, and soil. The exposure consists of increase in intensity of light, especially its ultraviolet fraction, frequent sudden variation in illumination through clouds forming and being lifted, and heavier winds. The soil is in both cases acid because of dampness and wet moss, and the dwarfing of the woody plants is mostly due to lack of deep weathered soil. The stunted, very slow growth on narrow crests is largely caused by starvation under humid conditions, and is ecologically comparable to the production of the bonsai miniature trees by the Japanese. If on such ridges there is a broadened depression with deep soil, the same trees and shrubs have a normal appearance.

However, no species of microtherm genera (genera of which all species are adapted to cold) are found on the low peaks just mentioned. Therefore the floristic zonation must be due to some other intrinsic ecological factor. There are good reasons to assume that this is a sharply limited response to low temperature by microtherm species; the limitation may be shared by all species of a genus. Within the tropics there is almost no seasonal variation in temperature but only diurnal variation. The temperature regime is therefore very regular. In the Malaysian lowland the daily average at sea-level is about 26·3°C, decreasing by 0·6°C for every 100 m elevation to 2000 m, and above 2000 m by 0·5°C per 100 m. The floristic zones mentioned above, tropical, montane, subalpine, and alpine, must therefore be considered to be due to a temperature response peculiar to the species. This has also been shown by transplantation experiments. If, for example, the cowslip (*Primula veris*) is grown at 2400 m in the scrub forest, it grows well, and flowers and fruits profusely; if it is transferred to about 1400 m it grows only vegetatively; but when transplanted to the original site it immediately starts flowering and fruiting. I have summarized [2] a long series of experiments by J. E. Teysmann in the years 1835–50 and later by H. N. Ridley and A. Rant. These experiments were mainly on European vegetables and ornamentals, but some were on native plants. They showed that below a critical altitude, fertility decreases first, and still lower down, vegetative growth also, each species having its own response. At 1000 m the hop (*Humulus lupulus*) refused to produce shoots, but if cuttings were kept in a refrigerator for 48 hours, vegetative growth was resumed for a while, but later again arrested.

FROST AND HARDINESS

Frost in the tropical mountains has sometimes damaged tea and coffee at altitudes as low as

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1500 m. The occurrence of frost depends on three factors: weather, vegetation, and topography. If days are cloudy, preventing warming by the sun, and if they alternate with cloudless and windless starlit nights, cold air caused by eradiation is likely to be preserved on the highlands and cooled further. This has no effect in high forest, because the cold air produced by radiation loss round the exposed surface of the leaves immediately sinks and is mixed with a thick layer of much warmer air below the canopy. If, however, the vegetation is very low, for example a short turf, the cooled air collects in a very thin sheet on the soil. If, moreover, the topography is level, or, more markedly, if it is a shallow hollow preventing the cool air from flowing off, the conditions are ideal for real frost. Just before sunrise, under such conditions, we have found a temperature of $-10°C$ a few cm above the soil, at 2000 m altitude. It is most unexpected to see tropical grasslands white with hoarfrost at so low an altitude. It is obvious that such frost sites ('frost pockets') are local. Silted up ancient crater lakes, or deforested plateaus suffer from it most, usually in the driest months of the year, which is when cloudless nights occur. It is clear that several factors must co-operate. If the hollows are shallow, the cold air flows downwards over the lowest part of the rim. In some places in Java, narrow deep draining trenches are dug in tea gardens on sites regularly suffering from frost; this is considered to be more effective than laying nightly smoke screens. It is clear that if highland plateaus have once been cleared or have repeatedly been burned (for hunting purposes), they will suffer from frost that will prevent tree seedlings from coming up. Grazing by cattle, or by deer attracted by grassland, keeps the turf low and thus favours low temperatures and removes the few hardy seedlings. Once deforested, such flats remain clear of forest.

Though we have used the word temperate to define subalpine and alpine tropical plants, the diurnal variation in temperature under which they live all the year round is not at all comparable ecologically with the growth period in the summer and resting period in the winter of temperate plants at our latitude. Experience has taught that hardly any temperate-tropical plants are hardy in Europe.

EFFECT OF ELEVATION

The upper and lower limits of species, particularly the lower one, by no means coincide with topographical contour lines. From the centre of the altitudinal range where the species is permanently at home, continuous upward and downward dispersal of diaspores (fruits, seeds, and spores) will tend to widen its range. Only those which by chance arrive in a suitable spot will establish the species. The frost pockets imitate to some degree the ecology of higher altitudes. Other favourable places outside the permanent range are banks of mountain streams, screes along valleys with bare soil, and waterfalls; the lowest localities of mountain plants are indeed located in such restricted habitats. The number of specimens there is always small, and they often remain sterile. From this it can be inferred that these colonies are beyond the normal range and depend for their continuance on receiving diaspores from elsewhere. This, I believe, is the explanation of the singular 'elevation effect' found by C. A. Backer and D. F. van Slooten [4] in comparing altitudinal records on the Javanese volcanoes. They stated that the lowest localities of mountain plants are confined to the highest mountains. For example, *Albizzia lophantha*, a very common indigenous small acacia-like tree, occurs in Java mainly between 1800 and 3100 m altitude, but exclusively on mountains that are at least 2500 m high or are in massifs of which the highest peak attains 2500 m; on such mountains it may occasionally be found as low as 1100 m. This means that it is absent on mountains of 2400 m, although there should have been an available range of $2400 - 1100 = 1300$ m. I have verified this discrepancy for many other mountain plants. I have further found that the effect becomes smaller in proportion to the cold to which the plant is adapted: for *Lonicera javanica* it is 900 m, for *Primula prolifera* only 460 m. Obviously the temperature response of the latter species is more strict and does not allow it to descend to local favourable spots lower down. I have explained [5] the elevation effect by assuming that the zone of permanent establishment, the continuous source for the descent of diaspores on the higher mountains, is absent on the lower mountains (figure 1). The chances that diaspores would disperse from one mountain to another and would lead to successful establishment in the zone of temporary establishment are almost nil.

It follows that, as a mountain loses height through erosion and decay, it will lose its mountain species when the height of the highest summit comes below the critical lower contour of their permanent establishment.

Where there are dry seasons, the dryness being caused by trade winds, the critical lowest contour

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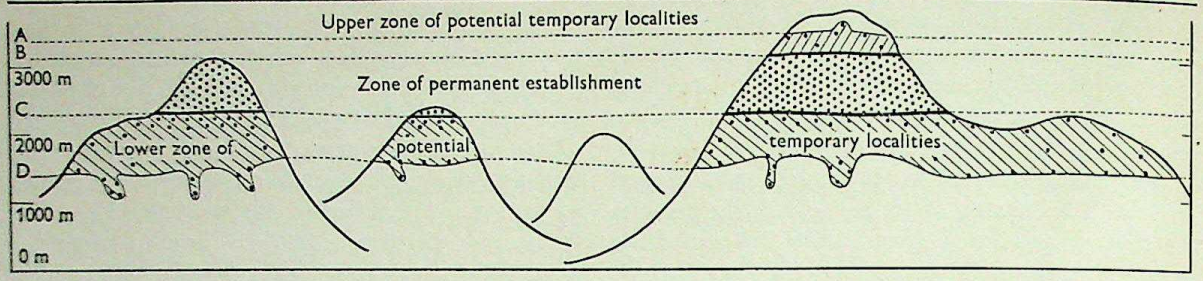

FIGURE 1 – *Schematic chart of an analysed altitudinal distribution of a mountain plant, illustrating the elevation effect. The density of dotting is in proportion to the number of individual localities.*

seems to be situated slightly lower than under permanently wet tropical conditions. In East Timor—this has dry seasons—the Portuguese have succeeded in growing peaches, apples, and plums in the mountains; this has not been possible in Java. The suggested explanation may be wrong, however, as success may have been due to the luck the Portuguese had in finding, by trial and error, strains capable of growing under short-day conditions; all our fruit trees are long-day plants. These conditions seem irrelevant to the native flora, as the areas of very many mountain plants extend from the tropics to, and even beyond, 40 degrees of latitude. This question should, however, be tested experimentally.

SOURCES OF THE MOUNTAIN FLORA

A point of prime interest is the origin of the tropical mountain flora. At all altitudes both woody and herbaceous plants are derived from two sources. Some are locally derived (autochthonous), some derived from plants that are not native to the area(allochthonous).

The species have been evolved *in situ* in genera that belong to plant families characteristic of the equatorial lowland forest. Most of the autochthonous families or genera, such as *Anonaceae*, *Bombacaceae*, *Dipterocarpaceae*, *Ficus*, *Meliaceae*, *Rhizophoraceae*, and *Marantaceae*, find their uppermost limit in the colline subzone below 1000 m, or at any rate fade rapidly away between 1000 and 2000 m. Only a few have some representatives at a much higher elevation than one would expect from their comparable latitudinal distribution, as for example species of the ginger family (*Zingiberaceae*) and of screwpines (*Pandanus*). There are many families, however, which have both a wide latitudinal distribution over the globe and a wide altitudinal distribution in the tropics, such as *Cruciferae*, *Labiatae*, *Oleaceae*, *Scrophulariaceae*, and *Theaceae*. Among them are also conifers, tree ferns, and genera familiar to botanists in temperate countries, who would expect them to occur only on tropical mountains; thus certain species of oaks and chestnuts, *Ajuga*, *Daphne*, *Podocarpus*, *Rhododendron*, and *Vaccinium* occur in Malaysia at sea-level. All these genera or families thus have a very wide ecology and can as a whole tolerate a wide range of temperatures; they have both heat-adapted (megatherm) and cold-adapted (microtherm) species. Their microtherm mountain species may have originated outside the tropics and migrated into them in the past, but it is also possible that they may have evolved *in situ* in the tropics from megatherm ancestors. For that reason they cannot serve as reliable material for solving the problem of how temperate genera have reached or crossed the tropics.

The allochthonous species form a minority in the tropical mountain flora, but they are of very great scientific interest, as they belong to genera, or sections of genera, of which all species are restricted to cold climates, and they give proof of it in the tropics where the lowest localities of their representatives are not found, even temporarily, below 1000 m altitude. This implies that they are found only on mountains at least 2000–2500 m high. *Androsace*, *Deschampsia*, *Gentiana*, *Primula*, *Valeriana*, and many others belong to strictly microtherm genera. They often cover a large area of distribution and may contain a very large number of species; yet none of the latter can tolerate a hot climate anywhere on the globe. The conclusions are that they originated as microtherm species and that their hereditary substance lacks the potentiality of adaptation to megatherm conditions. Thus they serve as thermometers of the past, and their distribution may tell us how they have reached or crossed the tropics from either the temperate zones of the northern hemisphere to those in the south, or *vice versa*.

ORIGIN OF THE MOUNTAIN FLORA

Charles Darwin and J. D. Hooker long ago

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concluded that such genera, in extending their range in the tropics, must have followed 'the most elevated routes'. In the New World, the Rockies–Andean chain provided a trans-tropical bridge; in Africa, the eastern highlands; and in Asia, the highlands of Malaysia. These bridges have of course had an extremely chequered paleogeographical history, and consequently the areas containing plant relics along such bridges must often show considerable gaps (figure 2). Still, several of such areas are reasonably continuous in Malaysia, as for example those of the genus *Euphrasia* and of *Haloragis micrantha* (figure 3). The chains show many 'stepping stones', indicating the course and function of the Sunda Bridge connecting the Asian and Australian continents.

It is apparent that the mineralogical composition of the bedrock that yielded the soil has relatively little importance; the same species is often found both on soils derived from igneous or sedimentary rock in the Himalayas and on volcanic soils in Sumatra and Java. But sometimes the antiquity of the rocks is reflected in the distribution of tropical subalpine and alpine plants. The subantarctic element, for example, is present in Malaysia only on high mountains of great antiquity, extending from New Zealand, Tasmania, and the Australian Alps to New Guinea, Ceram in the Moluccas, Central Celebes, Mt Kinabalu in North Borneo, Luzon, and North Sumatra. It does not occur on the high mountains of Timor or Java, which are as close to New Guinea and Australia as Mt Kinabalu, or even closer. By comparing distributional areas of species of the strictly microtherm Malaysian mountain plant genera,

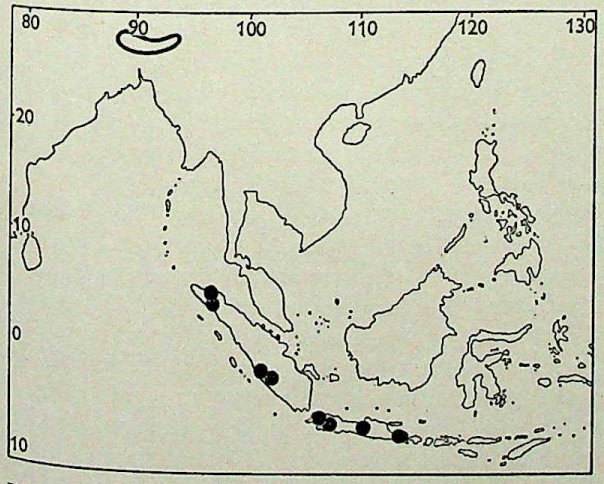

FIGURE 2 – *Localities of* Primula prolifera *Wall., showing a wide disjunction between North Sumatra and the Khasya Hills of Assam.*

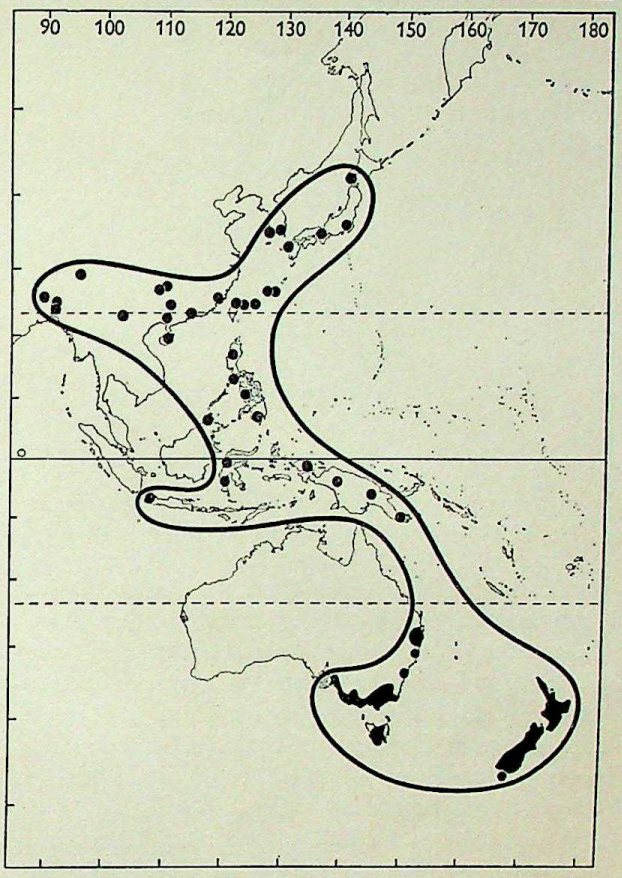

FIGURE 3 – *Distribution of* Haloragis micrantha *(Thumb.) R.Br., in the tropics. It is normally found at* 2000–3000 *m, occasionally descending to* 1600–1200 *m.*

about 900 species in all, it has been found that they fall clearly into three categories (figure 4). Mountain plants have followed three tracks in the dim geological past in extending their distribution over the tropical mountains, either from continental Asia towards Australia or *vice versa*; the plants seem to have migrated in plant communities and are still found growing together. This relation is so strict that I have repeatedly been able to predict approximately what category, and sometimes what genera, could be expected on unexplored mountains. In figure 4 the three main tracks along the trans-tropical Sunda Bridge have been indicated. It happens not infrequently that one species has followed two tracks, for example *Haloragis micrantha* (figure 3); *Anemone* has penetrated into the tropical mountains with two species passing along the Sumatran track and a third, belonging to a different section, independently along the Luzon–Formosa track. This has sometimes led to a remarkable intermingling of elements of different origin that became merged in a single plant community: Himalayan mountain herbs form most of

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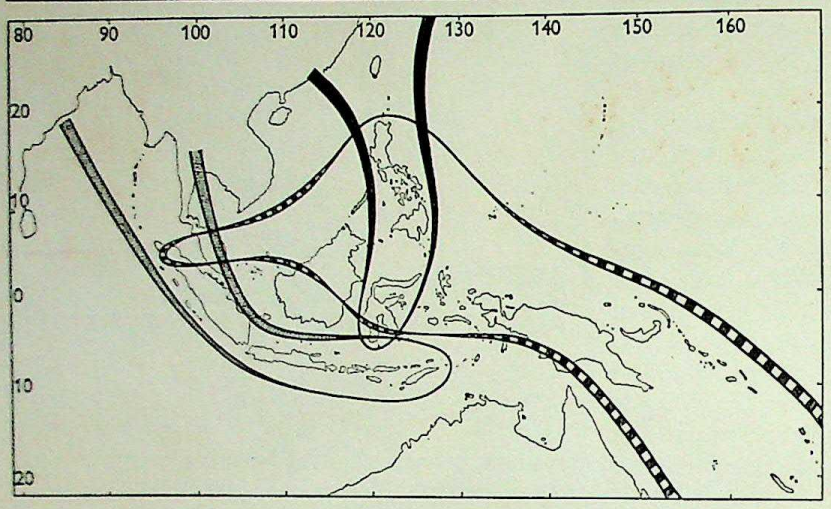

FIGURE 4 – *Tracks of microtherm genera of mountain plants in Malaysia.*

the undergrowth in the *Casuarina* forest of East Java and in the *Eucalyptus* forest on the Timor mountains, both *Casuarina* and *Eucalyptus* being distinctly of Australian origin. Conversely, the procumbent *Nertera* and *Pratia*, which stem from the sub-antarctic, form carpets in the subalpine ericaceous shrub-forest of West Malaysia, which is absent in the South Pacific.

The curious distributional regularities outlined above fall within the sphere of historical plant geography; they can only be properly elucidated in detail if the palogeographical history of the Sunda Bridge highlands after the Cretaceous is better known. It is of course almost certain that the mountain ranges have never been continuous at any one time, and that short-distance chance dispersal, bridging inevitable gaps, can apply to distances of from a few tens to at most a few hundreds of kilometres. The dispersal spectrum method first used by O. Stapf [3] has, however, clearly shown the fallacy of invoking long-distance dispersal, while the cooling effect of the Pleistocene Ice Age has in the tropics resulted only in lowering the upper floristic zones by about 400 m, the equivalent of about 2°C. This is corroborated by the comparison of the Tertiary and Pleistocene fossils of both plants and animals.

I have briefly outlined the past and present phytogeographical relations of the Malaysian mountain flora; *mutatis mutandis*, similar reasoning can be followed for the tropical mountain floras of Africa and of the New World.

COMMENT ON THE PLATES

The plates are reproductions of drawings made by Amir Hamzah and Mohamed Toha of plants that I collected; they were made for a detailed book on Javanese mountain plants whose publication was prevented by the outbreak of the second world war.

Plate I shows the seven *Rhododendron* species native to Java; among these *Rh. javanicum*, *Rh. retusum*, and *Rh. malayanum* were imported to Europe a century ago, mostly by Thomas Lobb, for the firm of Veitch. Here they were used for hybridization, together with the Malayan *Rh. jasminiflorum*. Except for *Rh. retusum*, *Rh. loerzingii*, and *Rh. zollingeri* they are almost always epiphytic shrubs. *Rh. album* was by error described as having white flowers, as expressed in its specific epithet; they are really sulphur-yellow with a few brown scales. Rhododendrons are a conspicuous constituent of the subalpine brushwood in Malaysia.

Plate II shows three kinds of *Begonia*, a group of lively-coloured herbs of the forest undergrowth. On the left are some curious plants, all parasitic and devoid of chlorophyll, belonging to the family *Balanophoraceae*. They possess an often very large, hard, subterranean corm attached to the root of the host tree from which break away short thick stems ending in an elongated or club-shaped spadix which is densely covered with thousands of tightly packed, minute flowers. In the *Balanophoras* the stems are provided with erect, stiff red (sometimes yellow) bracts. In the large *Rhopalocnemis* (left) the spadix is bractless but covered with tooth-shaped quickly falling scales; the lower flowers are male and protrude, the female flowers form one continuous velvety mass. In *Balanophora* the plants are usually either female or male and the corm is generally provided with pale star-shaped spots. Of the upper one (*B. globosa*), hitherto only female specimens have been found in Java, though males are known from the Malay Peninsula. The lower two belong to *B. elongata*, left a male, right a female. The sticky wax in the corms of *Balanophora* can be extracted by boiling and is used for making candles.

Plate III shows a few of the orchids in which the tropical mountains abound. On the left is a curious leafless, chlorophyll-lacking, pink-coloured saprophyte, a complete plant of *Galeola javanica*, found in deep shade on humus soil. It is half

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PLATE I – *Rhododendron species.* 1. Rh. javanicum (*Bl.*) *Benn.*, 2. Rh. citrinum *Hassk.*, 3. Rh. malayanum *Jack*, 4. Rh. loerzingii *J.J.S.*, 5. Rh. zollingeri *J.J.S.*, 6. Rh. retusum (*Bl.*) *Benn.*, 7. Rh. album *Bl.* (Plates I–IV, *slightly smaller than natural size.*)

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PLATE II – 1. Begonia longifolia *Bl.*, 2. B. lepida *Bl.*, 3. B. muricata *Bl.*, 4–5. Balanophora elongata *Bl.*, ♀ *and* ♂, 6. Rhopalocnemis phalloides *Jungh.*, 7. Balanophora globosa *Jungh.*

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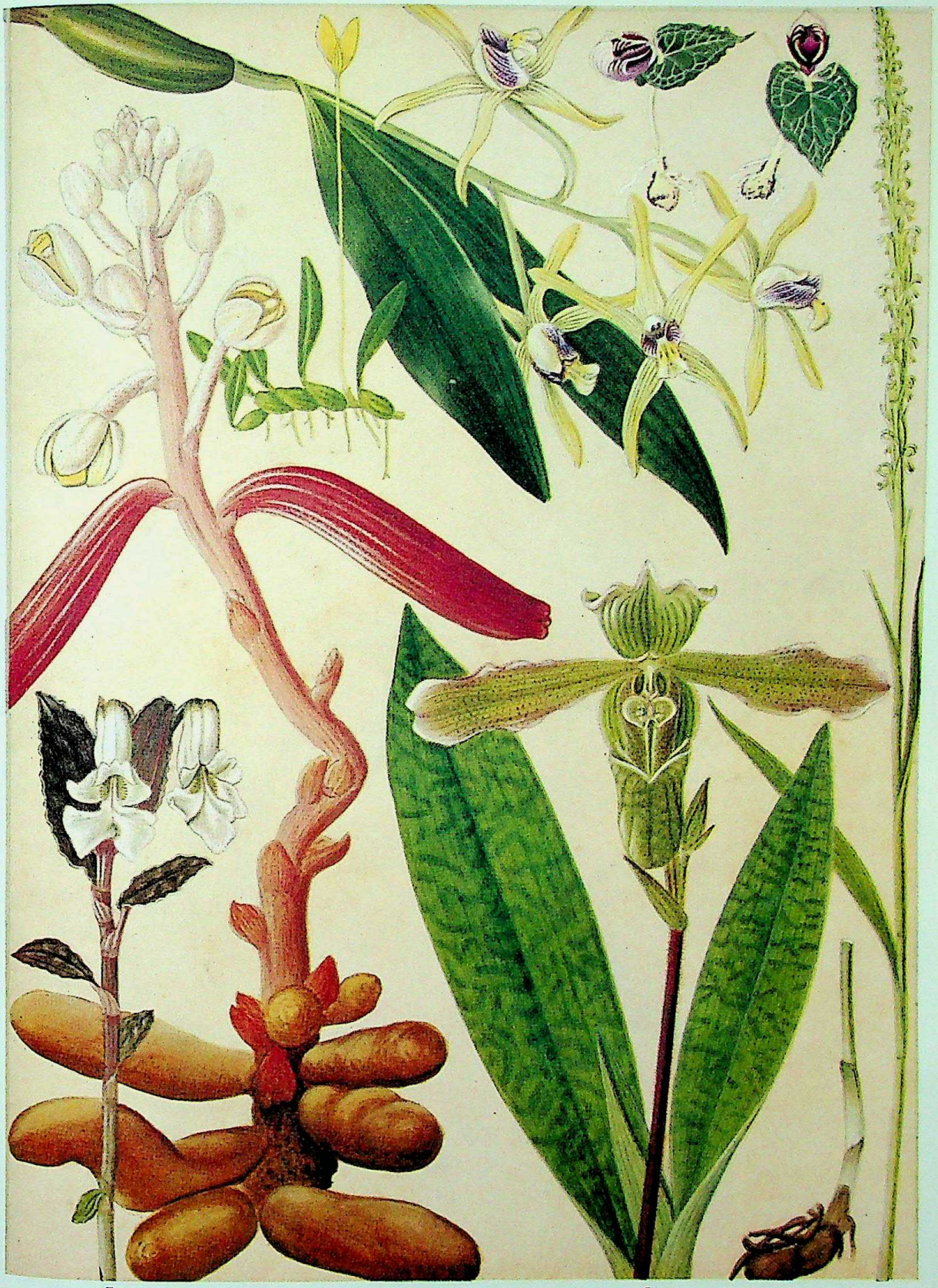

PLATE III – *Orchids*. 1. Dendrobium cymbidioides (*Bl.*) *Lindl.*, 2. Bulbophyllum ovalifolium (*Bl.*) *Lindl.*, 3. Corybas mucronatus (*Bl.*) *Schltr.*, 2 *specimens*, 4. Herminium lanceum (*Thunb. ex Sw.*) *Vuyk*, 5. Paphiopedilum javanicum (*Reinw. ex Lindl. and Paxt.*) *Pfitz.*, 6. Galeola javanica (*Bl.*) *B. & H.*, 7. Pristiglottis hasseltii (*Bl.*) *J.J.S. & Cretz.*

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PLATE IV – 1 *and* 9. Arisaema filiforme *Bl.*, *fruiting spadix and inflorescence*, 2. Orophea hexandra *Bl.*, 3. Euonymus japonicus *Thunb.*, 4. Arisaema barbatum *Buchet*, *spathe and spadix*, 5. Ilex spicata *Bl.*, 6. Schisandra elongata (*Bl.*) *Hook. f. and Th.*, 7. Rungia coerulea (*Bl.*) *Warb.*, 8. (*centre*) Arisaema inclusum *N.E.Br.*, *complete plant*, 10. Alyxia reinwardti *Bl.*, 11. Hypoxis aurea *Lour.*

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hidden in the ground and has very large tuberous roots and a few insignificant bracts. The lower two flowers have already set an almost fleshy pod; the yellowish flowers open only very little. In the other lower corner is one of the slipper orchids (*Paphiopedilum*). In the upper right corner two drawings represent the tiny, curious one-leaved *Corybas*, consisting of a single tuber, a single leaf, and a single large flower. It grows in moss, the leaf appressed to the substratum; after anthesis the pedicel becomes fleshy and lengthens considerably. The green orchid on the right, *Herminium lanceum*, is found in subalpine grassland.

In plate IV several families are represented, of which the three species of Jack-in-the-pulpit (*Arisaema*) with their remarkable hood-like spathe attract attention in the forest; the dark-coloured one (*A. filiforme*) is the largest and has a remarkably long tail-like appendage to the spadix; in the upper left corner is the spadix carrying the red berries. Left below is the liana, *Alyxia*, with white flowers and black berries; its bark is extremely tough; in drying it produces the fragrant cumarin. Upper left is one of the very few mountain species of the lowland family *Anonaceae*, a small tree with curious, mitre-shaped flowers, red fruits, and pale young leaves. Upper right is the attractive *Euonymus japonicus* which occurs both as an erect shrub in the open or as a root-climber in the forest. In the right lower corner is a liana with flowers and reddish berries, *Schisandra elongata*, belonging to the primitive Ranalian assemblage of the *Magnoliales*. Middle right is *Ilex spicata*. *Ilex* is a very common genus in tropical mountains; spiny leaves such as occur in the European holly (*Ilex aquifolium*) are an exception in the genus.

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Book reviews

SCIENTIFIC RESEARCH

Current Trends in Scientific Research, *by P. Auger. Pp.* 245. *Her Majesty's Stationery Office, London.* 1961. 33*s. net.*

The modest title of this book does not indicate the vast range of its contents. It covers not only pure research in all sciences but much of applied research and development, and it contains a good deal about the implications of the present impetus in scientific and technological advancement.

In general, the book achieves its objective of acting as a pointer to ways of developing better co-ordination of scientific knowledge and its effective dissemination. Obviously it has been impossible to achieve effective co-ordination in the space of 250 closely printed pages. It is, however, truly a survey and as such should be particularly valuable to the growing number of people, mainly scientists working in their own narrow field, who are asked for their advice and guidance in the wider issues of national and international science.

The book will not be easy reading for the interested layman. It is, however, an encouraging answer to the predatory critics who ask where science is heading. SIR HARRY MELVILLE

NUCLEAR MAGNETIC RESONANCE

An Introduction to the Analysis of Spin-Spin Splitting in High Resolution Nuclear Magnetic Resonance Spectra, *by J. D. Roberts. Pp.* 116. *W. A. Benjamin Inc., New York.* 1961. $4.95.

High-resolution nuclear resonance spectra of organic compounds are now used extensively for the elucidation of the structure of unknown substances and for the detailed study of related chemical problems. It is the purpose of this book to teach the organic chemist the underlying theory of these nuclear-resonance spectra and to show him how to use the more complicated cases to their greatest advantage.

The book starts off with a simple introduction describing the origins of chemical shifts and spin-spin coupling and the ways in which they affect the appearance of a high-resolution nuclear-resonance spectrum. The text is liberally provided with simple exercises which lead the reader step by step along the author's arguments. There are then a number of spectra that constitute problems for the reader to solve and that are of great help in familiarizing him with the situations he is likely to encounter. The rest of the book comprises a simple and clear account of the quantum-mechanical theory of spin-spin interaction between nuclei, starting with the simplest cases and leading finally to the so-called ABC and A_2X_2 systems. The arguments are extremely clear and, as in the first chapter, many exercises are given of gradually increasing difficulty which lead the reader along in a most satisfying way.

The book is not intended to be comprehensive, but to illustrate the principles that underlie the methods of analysing these more complicated spectra. As a first introduction to the subject, the book can be recommended most warmly to anyone interested in

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nuclear magnetic resonance. To the organic chemist in particular, this exposition can be of great help to a more complete understanding of the ways in which these spectra can be applied.

R. E. RICHARDS

ANALYTICAL CHEMISTRY

Treatise on Analytical Chemistry, *edited by I. M. Kolthoff and P. J. Elving, with the assistance of E. B. Sandell. Part II*, Analytical Chemistry of the Elements, *Vol. I. Pp. xxi+471. Interscience Publishers Inc., New York; Interscience Publishers Ltd, London.* 1961. *£6 net.*

The first volume of Part II of this ambitious work, edited by Professor Kolthoff and his two colleagues, comprises six main sections: Principles of Inorganic Nomenclature, by W. C. Fernelius; Determination of the Elements, General Concepts, by J. I. Hoffman; Hydrogen by H. F. Beeghly; Water, by J. Mitchell Jr.; The Inert Gases, by G. A. Cook; and The Alkali Metals, by S. Kallmann.

As with the other volumes in this series, each chapter bears the mark of authority. That by Dr Hoffman of the Bureau of Standards is more philosophical than factual in nature, but, containing as it does some timely advice to the young analyst, it is worthy of inclusion. In particular, the distinctions between analysis and measurement are brought out.

The chapter on hydrogen is restricted in the main to its determination in metals, in which connection Dr Beeghly is well known. The longest chapter in the book is devoted to water, which gives some indication of the complexity that attaches to the determination of this common substance: there are 356 references. This and the following chapter on inert gases deal with matters of great contemporary interest. As Dr Cook points out, the changes in the methods for the determination of the inert gases have been among the most revolutionary in analysis in the last decade. Indeed, techniques of gas analysis have until very recently lagged far behind those of nearly every other branch of the subject.

The final chapter, on the alkali metals, is of the same high standard as the remainder of the book and the 471 references cover work up to 1960.

This volume is well up to the standard of the other parts of the treatise and can be recommended without reservation to the professional analyst.

R. C. CHIRNSIDE

A Textbook of Quantitative Inorganic Analysis, including Elementary Instrumental Analysis (*third edition*), *by A. I. Vogel. Pp. xxx+1216. Longmans, Green & Co. Ltd, London.* 1961. *70s. net.*

Nearly twice the size of the first edition, the third edition of this now familiar text has been expanded rather than revised, particularly on the instrumental side, to include several useful techniques in its compendious and inevitably somewhat uncritical treatment. Theory receives attention throughout, often superficially, sometimes tersely to the point of uselessness. Copious use of commercial description, interesting to some, is pointless when trade literature is freely available, and theory stops short of the treatment requisite for intelligent reasoned choice of instrument. Modern and obsolete mingle: at least one instrument described did not reach production until after publication, others are relics; methods too new for general acceptance are included with others dimly remembered. Normalities are perpetuated, radiant powers and absorbances receive but passing mention. Titrimetric conversion factors, a useful check to routine analysis, are bad for students. The university syllabus coextensive with this book is Utopian; and the preoccupation with a mass of practical detail and the lack of emphasis on theory and principles do not commend this book for teaching. The practising analyst with the theoretical knowledge and practical experience necessary for selection and modification will find that the compendious treatment gives either the detail or the lead he seeks.

E. BISHOP

GAS CHROMATOGRAPHY

Gas Chromatography, *by D. Ambrose and Barbara A. Ambrose. Pp. vii+220. George Newnes Ltd, London.* 1961. *40s. net.*

In recent years, gas-chromatographic techniques have been applied to compounds previously thought too involatile, such as sterols, amino acids (as volatile derivatives), alkaloids, bile acids. These advances have been largely due to the development of the highly sensitive ionization-detectors. Since the technique has been taken up so widely, it is necessary to have a good introductory textbook.

Dr and Mrs Ambrose are to be congratulated on having produced a clearly written well-illustrated book that covers all the major aspects of the technique—column construction and operation, loading, detectors, retention volumes and their controlling factors, column performance, operation of high-efficiency columns, selective solubility, qualitative and quantitative analysis, gas analysis, preparative columns, and gas-solid chromatography. A small but adequate combined subject and author index is provided. It is unfortunate that the book was so slow in appearing that capillary columns and ionization detectors were not given as much space as they deserve. Nevertheless, the book can be confidently recommended to any beginner in the subject.

A. T. JAMES

PHARMACEUTICAL ANALYSIS

Pharmaceutical Analysis, *edited by T. Higuchi and E. Brochmann-Hanssen. Pp. ix+854. Interscience Publishers Inc., New York; Interscience Publishers Ltd, London.* 1961. $28.50.

The object of this volume as set out in the preface is to describe a collection of selected methods currently used in the laboratories of some leading pharmaceutical companies. The approach to the subject has been systematic, the book being divided into eleven chapters, each dealing with a group of structurally related drugs. Individual chapters are written by an expert in the field, and a common pattern of presentation is adopted throughout. A brief introduction is followed by a description of analytical methods applicable as far as possible to all members of the group. Specialized techniques applicable to selected members of the class then follow, and the chapter is concluded with a comprehensive and up to date list of references. The methods included range widely from classical gravimetric, volumetric, and colorimetric procedures to more specialized instrumental techniques employing ultra-violet and infra-red spectroscopy, fluorimetry, polarography, and chromatography. Microbiological methods are specifically excluded from consideration.

The editors have given predominant attention to drugs of a synthetic or microbiological origin, and with the exception of the alkaloidal group, which has been given extensive treatment, have made no attempt to describe assay procedures for the older vegetable drugs, many of which are fast disappearing from the pharmacopœas.

A. HOLBROOK

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ORGANIC FLUORINE COMPOUNDS

Chemistry of Organic Fluorine Compounds, *by M. Hudlický. Pp.* 536. *Pergamon Press Ltd, Oxford.* 1961. 60*s. net.*

This book was first produced in Czechoslovakia a few years ago. Its translation into English has been accompanied by a revision of the text and the incorporation of new work and references up to the end of 1959. The author has worked in this field for many years and has included results from his own researches.

The book emphasizes practical aspects of the subject. A brief introduction on nomenclature and the handling of fluorine compounds leads to a chapter on apparatus and materials. Fluorinating agents and the preparation of fluorine compounds are then dealt with in detail. There follows a comprehensive chapter on reactions. Treatment of the properties, analysis, and practical applications of these substances completes the book. Thus the coverage is wide, but the author has refrained from using theoretical considerations as a foundation for even a part of the book, so that overall it presents an untidy image.

None the less, the wide range of selected material, including over 1000 references to the original literature, will be useful to those working in this field. Author, subject, and compound formulae indices will make for easy reference.

E. J. FORBES

ORGANIC NITROGEN COMPOUNDS

Diazo and Azo Chemistry: Aliphatic and Aromatic Compounds, *by H. Zollinger (translated by H. E. Nursten). Pp.* 444. *Interscience Publishers Inc., New York; Interscience Publishers Ltd, London.* 1961. £6 4*s. net.*

In this volume the author has brought together two important groups of nitrogen compounds, the diazoalkanes and the aromatic diazo compounds. Much of the aromatic part has been taken from the author's earlier monograph *Chemie der Azofarbstoffe*, and has been thoroughly revised and brought up to date. The review of diazoalkane chemistry is most timely, for although aliphatic azo compounds have been known for over a century, the systematic discussion of their chemistry has perforce awaited the recognition of the general principles of radical reactions. In the present volume the author's main concern is with structure and mechanism; his approach is by discussion of physico-chemical relationships. The author has given particular prominence to modern work, and earlier studies such as have found a place in most standard textbooks are omitted. He has included a discussion of the Sandmeyer reactions while omitting matters of dyestuffs technology and dyeing, and in general the appeal is to chemists as a whole rather than to the dye specialist alone. The writing is clear, the subject matter full of interest, and the printing excellent. A most useful and stimulating monograph.

W. BRADLEY

PHOSPHORUS CHEMISTRY

Phosphorus and its Compounds, *Vol. II*, Technology, Biological Functions and Applications, *edited by John R. van Wazer. Pp. xvi*+955–2046. *Interscience Publishers Inc., New York; Interscience Publishers Ltd, London.* 1961. £13 3*s. net.*

The first volume in this series gave a new dimension to phosphorus chemistry by treating it as a unified discipline subject to self-consistent rules. The present volume falls below this high standard to the extent that this approach is largely ignored. In addition, the excellent chapters on the biological and metabolic functions of phosphorus have, whatever the author may say, little in common with the main technological theme of the book. These criticisms apart, this is a work of great interest to all concerned with the commercial applications of phosphorus compounds. The section on technology is as complete as published data will allow. Excellent descriptions are given of the occurrence of phosphorus minerals and the manufacture of the element and its more important organic and inorganic compounds, the sulphides and halides curiously being excepted. The applications of phosphorus compounds in fertilizers, food technology, water treatment, and many other fields are well summarized in the final chapters. Together, the two volumes of this work form a definitive text in this important field.

A. F. CHILDS

AMINO ACIDS

Chemistry of the Amino Acids, *by Jesse P. Greenstein and Milton Winitz. Vol. I, pp. xxi*+760; *Vol. II, pp. xi*+763–1816; *Vol. III, pp. xiii*+1819–2872. *John Wiley & Sons, New York and London.* 1961. £40 3*s.* 6*d. net.*

Most monographs published in the sciences in which progress is rapid generally become out of date within a few years. The work under review belongs to a different and more uncommon category. While it covers most important aspects of the chemistry of amino acids, it is distinguished by a scholarly and thoughtful historical approach that produces a sense of perspective and balance that is lacking in most modern scientific publications. At the same time it deals with the most recent advances made in the amino-acid field, and one cannot fail to be impressed by the mature judgment that is apparent on almost every page. Both Dr Greenstein and Dr Winitz have been active on so many different aspects of the field they discuss that they are able to offer views based on personal experience. The style is generally pleasing, and the proofreading has been most carefully done.

The chapter dealing with the configuration of optically active centres impressed the reviewer most favourably. This constitutes a complete, critical, and stimulating review on optical activity of amino acids and related compounds. The third chapter, which also reflects the special interest of the authors, is an account of amino acids in nutrition, and deals with this important subject in a very stimulating manner. The physico-chemical aspects of amino-acid chemistry are competently dealt with, but the various chapters on this aspect of amino-acid chemistry are probably not quite as outstanding as some of the others.

The account of the interaction of metals with amino acids is most stimulating, as is the very detailed and informative review of the procedures used for the synthesis of peptides. There is a good and detailed account of chromatography in all its aspects; of sequential analysis of peptides; and of methods used for the resolution of amino acids.

The third volume deals with individual amino acids, and this detailed and informative review will remain for a long time a standard text for anyone working in the field of amino-acid chemistry or biochemistry.

A. NEUBERGER

CRYSTALLOGRAPHY

Chemical Crystallography. An Introduction to Optical and X-ray Methods (*second edition*), *by C. W. Bunn. Pp. xiii*+509. *Oxford University Press, London.* 1961. 60*s. net.*

In the first edition of this book, published in 1945, Dr Bunn produced an account of optical and X-ray crystallography that was remarkable for its

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lucidity and for its breadth of coverage. The second edition is similar to its predecessor and has all its good qualities. Where necessary the first nine chapters have been brought up to date. The additional information includes recent discoveries about crystal growth; new apparatus for collecting observations by X-ray diffraction; and new aids for structure determination by trial and error, such as the use of anomalous scattering for the determination of phases and absolute configuration, the use of intensity statistics, and the use of optical diffraction.

The immense advances in the technique of crystal structure analysis can be judged from Chapter x, 'Direct and Semi-direct Methods'. This is more than twice the length of the corresponding chapter in the first edition, which went under the title 'Electron Density Maps and Vector Maps'. Discussion of maps now includes difference syntheses and vector maps with anomalous scatterers. Also incorporated are sign relationships, ultimate refinement and tests for accuracy, molecular transforms, and methods of determining phase angles.

The book gives a readable account of the scope and nature of the application of X-ray crystallography to chemical problems; it is an excellent introduction for research students starting X-ray crystal structure analysis, and its readability and non-mathematical treatment make it interesting to undergraduates.

M. R. TRUTER

GEOLOGY

A Hole in the Bottom of the Sea, *by W. Bascom. Pp.* 352. *Weidenfeld and Nicolson, London.* 1961. 25*s. net.*

This is a reprint of a book with the same title published in the United States some months ago, but all the plates have been omitted. The first half of the book gives an account of our knowledge of the Earth with special reference to the ocean floor and the Mohorovičić discontinuity—the Moho. The second half describes the techniques of drilling at sea and discusses how they might be adapted to drill a hole in deep water.

The drilling of deep holes in the floor of the ocean can tell us many things. Comparatively shallow holes will give a complete section of the unconsolidated sediments; deeper ones will sample the lavas, and perhaps baked and consolidated sediments among them. A hole going 5 km beneath the ocean floor should reach the Moho and give us a sample of the mantle of the Earth.

The present book was written before the trial drilling described in a recent article in ENDEAVOUR (20, 188, 1961), but it does describe the methods that were used in that operation and that may be used in drilling a very deep hole.

The book is popular in the sense that it is interesting and easy to read, but it also contains many things that will interest specialists in marine geology and geophysics.

SIR EDWARD BULLARD

PALAEOCLIMATOLOGY

Descriptive Palaeoclimatology, *edited by A. E. M. Nairn. Pp. xi*+380. *Interscience Publishers Inc., New York; Interscience Publishers Ltd, London.* 1961. $11.

The most remarkable inferences concerning the past climates of the Earth were made in the last half of the nineteenth century by British geologists, who, in various papers to the Geological Society of London, described evidence of widespread sea-level glaciations in India, Australia, South America, and Africa in Permo-Carboniferous times. They had, in rocks of this age, found the tillites, varves, and striated pavements that in northern Europe had been the signs of the recent glaciations. In order to explain these and other palaeoclimatic data, Wegener proposed in 1912 that the continents of the southern hemisphere and India once formed a single continent (Gondwanaland), which was near the South Pole in Permo-Carboniferous times and had drifted apart. Continental drift, having been disregarded for a long time, has now been made respectable once more by the recent advances in palaeomagnetism. An urgent task is the re-evaluation of all the geological indicators of past climates to see whether the latitudes assigned by palaeomagnetism to the different continents in the various geological periods explain the seeming anomalies in the climates of the past, or whether gross changes in the planetary climatic zoning are required. This authoritative book (with fifteen contributors) provides an excellent up to date review of the methods, inferences, and difficulties of interpreting the geological record of palaeoclimates.

S. K. RUNCORN

PALAEOBOTANY

Studies in Paleobotany, *by H. N. Andrews, Jr. Pp. xii*+487. *John Wiley & Sons, New York and London.* 1961. 94*s. net.*

This book contains a great deal of information about the plant world of the past, with references to most of the important discoveries made during the last decade. It will be a most useful book of reference and will at the same time be comprehensible to students of both geology and botany. The author has adopted a rather unusual arrangement of the subject matter. After an introductory chapter, he deals in Chapters 3–7 with the early ferns, the Pteridosperms and then the Angiosperms, with an excellent account of some European and North American Tertiary floras. He uses this series of chapters to indicate the evolutionary sequence leading up to the Angiosperms. Then follow chapters on the Pteridophytes, fossil floras of the polar regions, the Bryophyta, Palaeozoic and Mesozoic floras, and introduction to palynology by C. J. Felix, with finally a short account of some palaeobotanical techniques.

This arrangement leads to some confusion. Information about the origin of seeds, for example, is to be found in Chapter 5, where Carboniferous ovules and the ovuliferous parts of Triassic Pteridosperms are described, and much later, in Chapter 13, comes a discussion of heterospory and the evolution of the Pteridosperm seed. This arrangement also confuses the geological sequence. starting with Devonian and Carboniferous ferns and Pteridosperms, the Angiosperms are brought in, followed by the Conifers, Lycopods, and Horsetails. To the reviewer's mind very little is gained by this, and continuity is lost.

The book is well written and, apart from the arrangement to which reference has been made, will be found to be a most useful introduction to the subject.

J. WALTON

ECOLOGY

Ecology of Inland Waters and Estuaries, *by G. K. Reid. Pp. xvi*+375. *Reinhold Publishing Corporation, New York; Chapman and Hall Ltd, London.* 1961. 72*s. net.*

This book sets out to present a dynamic view of the factors and processes operating in natural waters. The emphasis is on principles rather than on excessive description. The treatment is clear, thought at times the writing is a little pedantic. The first two parts deal with the origins of lakes, streams, and estuaries, the relation of their physical properties to their ontogeny, and the physical and chemical properties of

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water itself. This is followed by a section dealing with the habitat and the factors of the environment. The factors are taken in turn, and a full general treatment of them in their dynamic aspect is given. Full attention is paid to processes such as cycling, although little mention is made of the physiological importance of the nutrients. An account in rather general terms is then given of the plants and animals of these environments arranged taxonomically. The examples given are mainly drawn from the American continent, but this does not necessarily detract from their value. Finally, the closing section deals with the inter-relations of the organisms and their environment.

This comprehensive account of habitat, environmental factors, and organisms involves much of fundamental ecological principles. The logical and up to date treatment these receive makes the book valuable to the general ecologist as well as to the specialist limnologist. In particular, the interrelationships between the organisms themselves and that with the environment together with the emphasis on the dynamic nature of the problems and principles involved, makes this a very useful textbook. F. N. WHITEHEAD

GENETICS

British Medical Bulletin, *Vol. XVIII, No.* 1, *January* 1962. Genetics of Microorganisms. *Pp.* 88. *The Medical Department, The British Council, London.* 1962. 20*s. net.*

The union of genetics, microbiology, and biochemistry has been extremely fruitful. It has yielded, during the last twenty or so years, some of the most exciting and fundamental advances in biology. These facts are well reflected in the present volume.

Following a stimulating introduction by Sir Macfarlane Burnet, there are sixteen papers, mainly of the survey type, by well-chosen authorities. The range of topics is broad. It includes nucleic acids, gene-enzyme relationships, most aspects of bacterial and viral genetics, and a paper on microbial genetics and chemotherapy. The final paper, on microbial genetics and human genetics, discusses the possible application of the approaches of microbial genetics to the study of human heredity. The size of the volume has required very careful selection of material from a wide field, and in general the choice has been well made. Bacterial, viral, and physiological genetics are well represented. The only conspicuous omission is in fungal genetics; a paper devoted largely or exclusively to this subject would have added balance and historical perspective and would have justified the title.

Almost all of the papers will satisfy the specialist and be read with profit and pleasure by the non-specialist. The volume is welcome; it provides an excellent, up to date, and largely well-balanced account of a very important field. J. A. ROPER

CELL HEREDITY

Cell Heredity, *by Ruth Sager and Francis J. Ryan. Pp. xi*+411. *John Wiley & Sons, New York and London.* 1961. 60*s. net.*

The sub-title describes the book as 'an analysis of the mechanisms of heredity at the cellular level'. The analysis covers three dovetailed topics, the identification of genes, their replication and transmission, the mechanisms of their action; it ends with a discussion of the problems raised.

This textbook ranges over many aspects of genetics, but takes as its basis contemporary work on the gene rather than classical Mendelism and cytogenetics, which are, however, treated as integral parts of the discourse. Topics such as human cytogenetics, which cannot yet be conveniently harmonized with the cellular view of gene action, are generally omitted. The book is suitable for undergraduates but will be appreciated by graduates, as well as by those with an elementary knowledge of chemistry and biology who are prepared for fairly strenuous reading. Theory is based on clearly presented experimental evidence, and the reading of concentrated information is made easier by lucid English. The choice of tables, figures, and photographs is excellent, and each chapter concludes with a useful general bibliography. The work as a whole shows how the classical material of a subject may be presented, together with the most recent advances, as a coherent living whole. J. BOSS

BIOCHEMICAL TECHNIQUE

Partition of Cell Particles and Macromolecules, *by Per-Åke Albertsson. Pp.* 231. *John Wiley & Sons, London and New York; Almquist & Wiksell, Stockholm.* 1961. 56*s. net.*

This book deals with a largely new possibility of fractionating complex biochemical mixtures by partition between the two phases resulting from a mixture of two suitable polymers in aqueous solution. In a foreword, A. Tiselius metions that the book represents the author's Ph.D. dissertation at Uppsala, but the extent and level of the work described are unusually high for such a project.

Various mixtures of polymeric materials (such as dextrans, substituted dextrans, methyl celluloses, polyvinyl alcohol, and polyethylene glycols), ionic as well as non-ionic, have been studied, and phase diagrams are provided with other relevant practical information. A general theoretical treatment indicates that the larger the particle-size of the material being studied, the more one-sided is the distribution anticipated between the two phases, and that interfacial absorption is favoured by large particle-size and large interfacial tension. With these guiding principles, the partition of whole cells, viruses, microsomes, proteins, and nucleic acids has been studied experimentally for the various two-phase systems as a function of type of salt, salt concentration, pH, temperature, and material concentration. The concentration of viruses receives special mention, and highly encouraging results are reported. The possibilities of counter-current distribution are also mentioned. Perhaps surprisingly, little denaturation or inactivation of biologically active materials occurs, and the author considers that a protective mechanism may be involved.

In spite of the extensive work reported it is too early as yet to pronounce on this partition method, but undoubtedly the book lays sound foundations for applications by others.

P. JOHNSON

METABOLISM

Metabolic Pathways, *Vol. II* (*second edition of* Chemical Pathways of Metabolism), *edited by David M. Greenberg. Pp. xiii*+814. *Academic Press, New York and London.* 1961. £8 11*s.* 6*d. net.*

This second and final volume, written by fifteen contributors, covers the metabolism of amino acids and related compounds (P. P. Cohen and H. J. Sallach, D. M. Greenberg, E. Kun); the synthesis of proteins (I. D. Raacke); the metabolism of purines, pyrimidines, nucleosides, nucleotides, and polynucleotides (M. P. Schulman, L. Warren); of porphyrin pigments (S. Granick and D. Mauzerall); and of

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a number of B vitamins (P. Albersheim and J. Bonner, N. O. Kaplan, G. W. E. Plaut, T. H. Jukes, and H. P. Broquist). Degradation as well as the synthesis of these substances is considered. The work is a most competent, up to date, account of the present knowledge of metabolic pathways. Those of animal tissues and micro-organisms are treated comprehensively, with hardly any major gaps and with the fullest references to the literature. Plant metabolism is less fully covered. Though many aspects of plant metabolism are mentioned, others, such as the formation of alkaloids, rubber, lignin, and pectic substances, and certain aspects of photosynthesis, have been omitted. As an advanced reference book, the two volumes will be a very valuable guide to the subject of intermediary metabolism.

H. A. KREBS

ENZYMOLOGY

Advances in Enzymology, *Vol. XXIII, edited by F. F. Nord. Pp. v+557. Interscience Publishers Inc., New York; Interscience Publishers Ltd, London.* 1961. $15.50.

The long series of annual volumes of 'Advances in Enzymology' has established a high standard. Against this background, the present volume may appear slightly wanting, especially in the proportion of articles that may be expected to remain standard references on their subject matter for some time to come.

Two of the articles relate to particular groups of enzymes. Collagenases and elastases are discussed by I. Mandl, who makes a convincing case for the separate existence of these enzymes. P. Desnuelle gives an account of pancreatic lipase, to the understanding of which he and his co-workers have made so useful a contribution.

The volume contains two valuable articles on metabolism: by E. Racker, primarily on oxidative phosphorylation, and by H. L. Kornberg and S. R. Elsden on the bacterial metabolism of 2-carbon compounds. Two articles are of a chemical nature. These are a short account of the chemistry of mevalonic acid, by A. F. Wagner and K. Folkers, and a very important contribution by R. Lemberg on the cytochromes of Group A. This contains probably the best account to date of the complicated and somewhat controversial chemistry of the haem *a* prosthetic groups.

At the other extreme, two articles on periodic enzyme reactions and protein configurations from the viewpoint of internal rotation potential, while containing some interesting ideas, hardly seem to merit one-seventh of the space in an annual volume of this kind.

E. C. WEBB

HAEMATIN ENZYMES

Haematin Enzymes, *edited by J. E. Falk, R. Lemberg, and R. K. Morton. Part I, pp. xxii+362; Part II, Pp. xviii+363–666. Pergamon Press Ltd, Oxford.* 1961. *£8 net, the set.*

This book has been produced from the symposium of the International Union of Biochemistry in 1959, and covers the widest field, from quantum mechanics to microbiology.

Practically every aspect of haematin enzyme research of current interest has been touched on. There are several papers on the physico-chemical properties of the porphyrins and their metal complexes, and in particular on the electronic structure and transport of the metal ions and the investigation of metalloporphyrin spectra. There is detailed discussion of the mechanisms of haem binding to globin and other proteins, and the incorporation of iron into haemoglobin. A considerable section deals with the haematin enzymes relevant to microbiology, for example the cytochrome oxidases of *Pseudomonas aeruginosa* and the atypical haemoprotein of the purple photosynthetic bacteria. An important part of the symposium is devoted to the cytochromes, their spectra, their occurrence in plant tissues and microsomes, and their mechanism of action in the intracellular respiratory chain. There is a full record of the discussion which followed each paper.

A. GOLDBERG

NEUROPHYSICS

Sensory Communication, *edited by W. A. Rosenblith. Pp. xiv+844. The M.I.T. Press, Massachusetts Institute of Technology; John Wiley & Sons, New York and London.* 1961. *£6 8s. net.*

These forty-two contributions to a symposium held at M.I.T. in 1959 provide a fascinating survey of various approaches that are being used in the attempt to understand the nervous system. The influence of analogies with human communication systems is pervasive, but there is little detailed use of the mathematics of information theory. Rushton provides a useful discussion of the implications of the fact that nerves are 'cables' of salt solution with poor conductivity. Hallowell Davis gives an account of peripheral coding in the auditory system that is both clear and distinguished, and takes repeated looks at the whole problem. P. D. Wall gives an example in which study is proceeding beyond the first steps in coding, to the central analysers, cells in the spinal cord. These act as amplifiers, each serving a large area of skin by means of a converging system of bundles. An example of analysis at the highest level is that by R. Jung, who shows how single neurons of the visual cortex integrate impulses from the vestibular receptors and other systems, as well as from the eye.

Among this feast of information, it is the papers with specific data to relate that hold the attention most successfully. The nervous system is so variously and wonderfully made that we are ever gaining new glimpses into the individual actions of its various parts. Those psychologists and theoreticians who are brave enough to try to keep our thoughts on whole sections or the entire organism may find our interest distracted from them by the fascinating details of the performance of some one system. It is inevitably difficult to say much about the whole until we know more of the parts. This book provides a valuable sample of the sorts of fact that are available.

J. Z. YOUNG

PLANT RESPIRATION

Respiratory Metabolism in Plants, *by H. Beevers. Pp. xi+232. Row-Peterson & Co., Illinois and New York.* 1961. $3.85.

This is the second volume of the Row-Peterson Biological Monograph series and like its predecessor has been written by a well-known authority actively engaged in the field. The book is a review of approximately the last twenty years' work, and no attempt has been made to cover the whole of respiration, but only those biochemical events of possible importance in respiration that occur in higher plants.

The book is in two parts. The major part deals with the reaction pathways of the initial stages of glucose breakdown, TCA cycle, electron transport and oxidase systems, and the energetics of respiration and intracellular location of enzymes. The second part discusses the relationship of respiration to other processes, including growth, solute

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transfer, nitrogen metabolism, photosynthesis, and fat metabolism.

Professor Beevers's close contact with the experimental situation has enabled him to present the facts clearly and in an uncontroversial manner; the relationship of respiration to other processes is dealt with in less detail.

The book has been written with the needs of honours and graduate students in mind, and it fulfills the purpose admirably. It should also prove extremely useful to more experienced research workers, especially those in related fields who wish to have an up to date review of the subject. D. BOULTER

MAMMALIAN EGG

The Mammalian Egg, *by C. R. Austin. Pp. viii+183. Blackwell Scientific Publications, Oxford.* 1961. 42*s. net.*

This brief introduction to the cytology of the mammalian ovum, which is comprehensive and enlightening to a degree hard to believe attainable within the compass of 183 pages, is not just a compilation of existing knowledge of the egg. It is a deeply thoughtful and thought-provoking synthesis that will be read by all those workers who use the mammalian egg as an experimental object.

Too often in the past cytological studies of the egg have been primarily descriptive. In this study the approach is from the functional rather than the purely morphological standpoint, and much emphasis is placed on the method of experimentation. The book is not steeped in histochemistry; its true merit is that the real biology is there. Furthermore, Austin never forgets that it is the task of the biologist to solve biological problems, and the essence of this book is that he brings an acute mind to the study of the evidence. It is unlikely, however, that the book will be accepted uncritically by some specialists and laboratory workers. But the fact that biologists will disagree among themselves on many points means only that the subject is very much alive and that more research is necessary.

The last section of the book describes the manipulation of eggs. This is a restrained, reasonable account which forms an effective conclusion to a book that is outstanding for its clarity and simplicity throughout. There are many excellent diagrams to illustrate the text, and a comprehensive series of photographs, many in colour. This book is completely up to date with an extensive bibliography, and it can be recommended to all who wish for a better understanding of the subject.

E. C. AMOROSO

MILK SECRETION

Milk: The Mammary Gland and its Secretion, *edited by S. K. Kon and A. J. Courie. Vol. I, pp. xiii+315; Vol. II, pp. ix+423. Academic Press, New York and London,* 1961. *Vol. I,* 100*s. net; Vol. II,* 86*s. net.*

The first of these two volumes contains twelve reviews, by different authors, dealing with the structure and development of the mammary glands of both farm animals and humans, and with the physiology and biochemistry of lactation. The second volume has eight reviews dealing with the nutrition of lactating women and farm animals, and with the nutritional value of milk. This complete, detailed, and authoritative survey of the subject was badly needed.

Through the years, research on milk secretion has been stimulated by its importance in farming, but, as a result, research workers have often had lower critical standards than are usual among physiologists, and many original papers must be carefully scrutinized before one accepts their conclusions. This scrutinizing should be the task of reviewers, but many reviewers in the past have merely catalogued the often conflicting conclusions of research workers. The present volumes have been edited by workers at the National Institute for Research in Dairying, where critical standards have always been high. The reviews have the reliability that scientists normally expect. They draw their own conclusions from the literature, and these may generally be accepted without further checking. J. M. BARRY

NEUROLOGY

Experimental Neurology, *by Paul Glees. Pp. xii+532. Clarendon Press; Oxford University Press, London,* 1961. 75*s. net.*

This book is a translated and considerably revised version of the author's *Morphologie und Physiologie des Nervensystems,* which appeared in 1957. One must salute his heroic attempt to present basic neurology as a single subject instead of perpetuating the usual false dichotomy between the 'anatomical' and 'physiological' aspects of the nervous system. There can be no doubt that Professor Glees's approach to the subject is the only possible one in the second half of the twentieth century.

Despite the fact that references to the author's own work appear in every chapter, the treatment of different topics is very unequal. For example, the chapter on the cerebellum leaves a great deal to be desired, while that on the visual system could hardly be bettered in a book of this size. It is good to see a chapter of twenty pages devoted to the reticular system, but many workers in the field would disagree with the relative emphasis, or lack of it, laid on various aspects of this important subject. In general, it is difficult, save for the expert, to disentangle established fact from hypothesis, for the distinction is not always clearly made in the presentation.

The book is excellently produced and excellently translated; but one cannot help wondering at whom it is aimed, for the detail is insufficient to make it a reference work for the researcher, while the text is too long for the undergraduate student. However, the detailed, though incomplete, bibliography to every chapter makes it a useful starting-point for almost any topic in the field of basic neurology.

DAVID BOWSHER

PROTOZOA

An Introduction to the Study of Protozoa, *by Doris L. Mackinnon and R. S. J. Hawes. Pp. xix+506. Clarendon Press; Oxford University Press, London.* 1961. 63*s. net.*

This is a good solid book on Protozoa, along modernized old-fashioned lines. It describes almost all the Protozoa, with the deliberate exception of the malaria parasites, that a university student is likely to encounter in class material. It also gives a sensible account of the principles and practice of microscopy, which first-year students should study, and some useful directions on the culture of Protozoa and on their preparation for microscopical examination. I recommend it for university practical classes in preference to any other book.

The book is arranged in accordance with the classification of the Protozoa, and this presents the authors with the opportunity for modern taxonomy, but inhibits serious physiological treatment. More also could have been made of the results of electron microscopy, which should be continually brought before students for interpretation.

Stories of encystment in *Amoeba proteus* are treated with firm disbelief, and presumably Leonardi's (1958)

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claim of gametes for *A. spumosa* would have been similarly dismissed, had it received attention. Various other omissions do not detract seriously from the usefulness of the book.

J. A. KITCHING

HISTORY OF SCIENCE

The Mechanization of the World Picture, *by E. J. Dijksterhuis (translated by C. Dikshoorn). Pp. viii+539. Clarendon Press, Oxford.* 1961. *£5 5s. net.*

The explanation of Being and Becoming by the Greek Atomists in terms of the motion of atoms in the void has, since its resurrection in the sixteenth and seventeenth centuries, been uniformly successful in 'explaining' an enormous and ever-increasing range of natural phenomena. Professor Dijksterhuis' work *De Mechanisering van het Wereldbeeld*, of which an excellent English translation has now been supplied by Miss C. Dikshoorn, traces the chequered history of this notion of mechanism in physical science from Greek times to the appearance of Newton's *Principia*. At the same time it supplies a history of the exact sciences, excluding mathematics, over the same period, unrivalled, within the compass of a single volume, as regards completeness, breadth of treatment, and mastery of significant detail. Specific criticisms can naturally be levelled against so extensive a work. For example, it could be argued, I think conclusively, that Newton, in the *Principia*, did not hold to the impetus theory imputed to him in paragraph 295. Also the present epilogue seems to me hardly adequate as it stands. Such criticisms, however, point to no more than minor blemishes in this magnificent book, which will inevitably long remain the standard work on the subject.

J. W. HERIVEL

THE NEWTON LETTERS

The Correspondence of Isaac Newton, *Vol. III*, 1688–1694. *edited by H. W. Turnbull. Pp. xviii+445. Published for the Royal Society by the Cambridge University Press, London.* 1961. *£7 7s. net.*

This magnificent edition of the Newton correspondence has now reached the period immediately following the publication of the *Principia*, and many of the texts here printed record the characteristic reactions of Newton's contemporaries to the implications of that historic book. Huygens has to be supplied with fuller information about resisted motion and the speed of outflow of liquids under pressure. To Richard Bentley, preparing his Boyle Lectures against atheism, Newton addresses his four cosmological letters. Memoranda on motion and gravitation, drawn up by Newton in his Woolsthorpe days and never before published in full, reveal a hitherto unsuspected link with the work of Galileo. And in letters to Wallis and jottings by David Gregory the doctrine of fluxions begins to be unfolded. In these pages, too, we can trace the beginning of Newton's friendship with John Locke and the first rumblings of his quarrel with Flamsteed. Other letters reveal him in the roles of biblical critic, chemist, and member of the Convention Parliament.

This edition is designed to include all known letters written by or addressed to Newton together with extracts from his memoranda and from contemporary writings. English translations are appended to Latin texts, and technicalities and historical references are elucidated. The illustrations include a portrait by Jervas and six facsimiles of manuscript. Despite the lamented deaths of the editor, Professor H. W. Turnbull, F.R.S., and of his associate, Mr. H. W. Robinson, there are good hopes that this great enterprise will go forward. The further volumes will be eagerly awaited.

A. ARMITAGE

MAX VON LAUE

Max von Laue. Gesammelte Schriften und Vorträge, *edited by M. Kohler. Vol. I, pp. xx+548; vol. II, pp. xlv+513; vol. III, pp. xliv+265. Friedrich Vieweg & Sohn, Braunschweig.* 1961. *DM* 145.

Max von Laue, the doyen of German physicists, died in April 1960 in his 81st year. It is highly creditable both to the publisher and to the editor, Professor Kohler, long a collaborator of von Laue, that the three volumes of collected papers and discourses became available with so little delay. The selection of papers was simplified by the fact that Laue himself drew up a list of the papers that he considered suitable to appear in a selection of his writings, and wrote an introductory note, 'Personal Reflections on my Work', dated February 1960, that prefaces the collection.

The first two volumes contain 91 of Laue's original papers, mainly on the topics of optics and thermodynamics, relativity, X-ray diffraction, the electron gas in electronic tubes, and superconductivity, together with other, more occasional, subjects. The coverage of Laue's production is fairly complete. But even if, given the chance, Laue might have included in a revised list some of the further papers that one would expect to find, such omissions are not serious in the fields that are covered by his books, namely relativity, X-ray and electron diffraction, and superconductivity. The original papers themselves show, of course, more of the motivation and genesis of his work than appears in the books.

Volume III should appeal to a wide readership. It begins with a 34-page autobiographical essay of Laue, which was published in 1952. Like most autobiographies of critically minded personalities, this is of great interest to read, especially as Laue was a master of precise prose. The same holds for the forty selected essays, speeches, and obituary notices that fill the remaining 265 pages of this volume. All are written with a deep sense of obligation to historical development and personal achievement. It is in this volume that Laue's personality finds its fullest expression.

P. P. EWALD

HISTORY OF MEDICINE

Medicine and the Navy, *Vol III*, 1714–1815, *by C. Lloyd and J. L. S. Coulter. pp. xi+402. E. & S. Livingstone Ltd, Edinburgh and London.* 1961. *50s. net.*

This book will be welcomed by all familiar with Volumes I and II, which were written by the late Surgeon-Commander John Keevil. The authors of the present volume have made one modification in plan, and instead of the chronological arrangement of the earlier volumes have written sections on the four following subjects: the medical department; the medical history of the wars of the eighteenth century; the naval hospitals; and sea diseases. These are followed by appendices and indexes to manuscript and printed sources, and index.

This scholarly work presents a wealth of information in an attractive manner. It should appeal not only to medical and pharmaceutical historians but to all who are interested in the social life of the period. Many of the developments which took place during the period under review had an influence that extended outside naval circles. For example, Dr Lind's work on scurvy was a landmark in the history of dietetics, and the improvements made in the naval hospitals were reflected in the planning and administration of civilian ones.

G. E. TREASE

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Some books received

(*Note*. Mention of a book on this page does not preclude subsequent review.)

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Die Übersetzung naturwissenschaftlicher und technischer Literatur, *Vol I, by R. W. Jumpelt. Pp. xv+214. Langenscheidt KG, Berlin-Schöneberg.* 1962. *DM.* 24.50.

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The Theory of Graphs and its Applications, *by Claude Berge (translated by Alison Doig). Pp. x+247. Methuen & Co. Ltd, London.* 1962. 36*s. net.*

Group Theory, *by Morton Hamermesh. Pp. xv+509. Addison-Wesley Publishing Co. Inc., Reading, Mass. and London.* 1962. 113*s. net.*

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The Fascinating World of Astronomy, *by Robert S. Richardson. Pp. viii+274. Faber and Faber, London.* 1962. 25*s. net.*

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Classical Electricity and Magnetism, *by W. K. H. Panofsky and M. Phillips (second edition). Pp. xiv+494. Addison-Wesley Publishing Co. Ltd, Reading, Mass. and London.* 1962. 72*s. net.*

Concise Physics for Ancillary Degree Students, *by R. B. Morrison. Pp. xii+539. Edward Arnold (Publishers) Ltd, London.* 1962. 35*s. net.*

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The Many-body Problem, *by D. Pines* (Frontiers in Physics *series*). *Pp. xii+457. W. A. Benjamin Inc., New York.* 1962. $3.95.

Physics and Techniques of Electron Tubes, *Vol. I, by R. Champeix. Pp. xii+221. Pergamon Press Ltd, Oxford.* 60*s. net.*

Réalisation d'un périodographe optique—Applications à l'étude des fonctions de transparence, *by Jean Ch. Viénot. Pp. 140. Service de Documentation et d'Information Technique de l'Aéronautique, Paris.* 1962.

Tables of Constants and Numerical Data, *vol. 12, by P. Aigrain and M. Balkanski. Pp. 65. Pergamon Press Ltd, Oxford.* 42*s. net.*

An Introduction to the Physics of Vibrations and Waves, *by N. Feather. Pp. viii+330. Thomas Nelson & Sons Ltd, Edinburgh.* 1962. 30*s. net.*

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Collection of Problems in Physical Chemistry, *by J. Bares, C. Cerny, V. Fried, and J. Pick (translated by H. Watney). Pp. xviii+608. Pergamon Press Ltd, Oxford.* 1962. 70*s. net.*

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Quantitative Chemical Analysis and Inorganic Preparations, *by R. M. Caven (second edition). Pp. 428. Blackie & Son Ltd, London and Glasgow.* 1962. 35*s. net.*

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Biology: An Introduction to the Science of Life, *by C. J. Goodnight, M. L. Goodnight, and R. R. Armacost. Pp. vii+460. John Wiley & Sons Ltd, London and New York.* 1962. 53*s.*

Intermediate Biology, *by W. F. Wheeler (sixth edition). Pp. xv+700. Heinemann Educational Books Ltd, London, Melbourne, Toronto.* 1962. 30*s.*

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Vade-mecum des antibiotiques et agents chimiothérapiques anti-infectieux, *by M. Neuman. Pp. 410. G. Doin & Cie, Paris.* 1962.

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Plant Life of Palestine, *by M. Zohary. Pp. vi+262. The Ronald Press Co., New York.* 1962. $8.

Plants—An Introduction to Modern Botany, *by V. A. Greulach and J. Edison Adams. Pp. xvi+557. John Wiley & Sons Ltd, London and New York.* 1962. 57*s. net.*

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Handbook for Telescope Making, *by N. E. Howard. Pp. x+322. Faber and Faber. London.* 1962. 45*s. net.*

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Rocket Engines, *by S. L. Bragg. Pp. 158. George Newnes Ltd, London.* 1962. 35*s. net.*

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Michel Eugene Chevreul, *by Albert B. Costa. Pp. 116. The State Historical Society of Wisconsin, Wis.* 1962. $3.50.

De Telescopio, *by Giovan Battista Della Porta. Introduction by Vasco Ronchi and M. A. Naldoni. Pp. 163. Leo S. Olschki, Florence.* 1962. 3000 lire.

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